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DEPARTMENT OF COMMERCE

UNITED STATES BUREAU OF MINES
SCOTT TURNER, DIRECTOR

## INFORMATION CIRCULAR

# RADIUM

BY

PAUL M. TYLER

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DEPARTMENT OF COMMERCE--BUREAU OF MINES

# RADIUM[1]

By Paul M. Tyler[2]

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2. Chief engineer, rare metals and nonmetals division, U. S. Bureau of Mines, Miss E. P. Youngman assisted in arranging and editing the material.

## FOREWORD

The literature on radium is already voluminous, but mcuh of it is too technical to be intelligible to the average reader, and there is no single publication that covers certain economic features of the radium industry. As a result of the renewed interest in the possibilities of domestic carnotite deposits a number of requests have been made to the Bureau of Mines for information, more particularly with reference to the business aspects of the industry. In order to meet this apparent need, the present paper has been prepared from material available in the files of the bureau. The technical data are compiled from various sources and are believed to be reasonably dependable, but the attempt has been not so much to achieve meticulous scientific accuracy as to bring together facts of industrial significance and popular interest.

## GENERAL DESCRIPTION

Radium is a brilliant white metal that melts sharply at about 700°C. and volatilizes appreciably at that temperature. With an estimated specific gravity of nearly 6.0, it is more than twice as heavy as aluminum and 25 per cent lighter than iron. Although it has been isolated in its metallic state, it is not used in that form, partly because it tarnishes rapidly even in dry air (forming a nitride), partly because of the greater expense of reducing it to metal, and partly because of the difficulty of dealing with such minute quantities of material. It is used always in the form of its compounds, the most important of which from the viewpoint of utilization are the bromide, chloride, and sulphate.

Radium compounds are similar in many ways to those of the alkaline-earth metals. With barium, in fact, it forms isomorphous salts which can be separated only by virtue of their difference in solubility. The physical and chemical properties of the compounds of the alkali and alkali-earth metals usually show a well-marked gradation in the order: calcium, lithium, strontium, barium, sodium, potassium, rubidium, and caesium; and in this scheme radium comes between barium and sodium. In so far as the volatility of the element is concerned, however, radium resembles calcium rather than barium.[3]

The oustanding property of radium is its radioactivity, which it imparts also to its salts. Radium metal decomposes water vigorously, evolving hydrogen and forming hydroxide in solution. The chloride, bromide, nitrate, carbonate, sulphate, azoimide, and platino cyanide of radium are known. When freshly prepared all of these are colorless, but they develop color on standing. Radium chloride closely resembles barium chloride. It is a soluble, -white crystalline salt, paramagnetic, and hygroscopic. The bromide is an unstable substance, losing bromine when kept, and passing into the hydroxide. It is a white crystalline salt, containing, when pure, 53.6 per cent of the element. Radium sulphate is the least soluble of all the sulphates.

Radium appears on the market usually in the form of anhydrous bromide mixed with more or less barium bromide and packed in small sealed glass tubes.

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[3] Thorpe, E., Dictionary of Applied Chemistry. Vol. 5, 1924, p. 640.

If freshly sealed, both the glass and contents are colorless or nearly so, but after some time the tubes are likely to become a deep violet color and the salt is likely to turn to a brownish tinge. Especially in freshly prepared tubes the salt may be luminous in the dark, but this is due to the presence of minor impurities, as neither radium nor its compounds are luminous. The glass tubes are usally encased in lead as a protection to those that must handle them.

## RADIUM STANDARD

The international radium standard was prepared by Madame Curie and deposited at Paris, but duplicates of the original have been placed in the leading capitals of the world. Radium preparations are now measured by comparing the electrical energy carried by the gamma rays against those from the standard. In the United States, radium is certified by the Bureau of Standards by comparing with a duplicate standard sample obtained in 1914.

## RADIOACTIVITY

The term "radioactivity" is now generally applied to a class of substances—such as uranium, thorium, radium, and their compounds—that possess the property of spontaneously emitting radiations capable of passing through plates of metals and other substances opaque to ordinary light. The characteristic properties of these radiations, in addition to their penetrating power, are their action on photographic plates and their power of discharging electrified bodies—for example, an electroscope. Also, a strong radioactive body like radium is able to cause marked phosphorescence and fluorescence in certain substances placed near it, and it will ionize gases. The spontaneous generation of heat is still another characteristic property of radioactive substances.

These phenomena may perhaps be better understood if radio activity is described as the result of the explosion of atoms. In a single gram of uranium, 5,000 atoms break down each second. Nevertheless, among radioactive substances uranium is classed as a relatively sluggish element; it takes five billion years for even one-half of a given piece of uranium to dissipate itself spontaneously. Radium breaks down far more rapidly; but despite the large number of its atoms that break down each second, its activity is maintained almost undiminished for an astonishing period of time. The half life of radium is reported as 1,690 years.[4] The atoms, when they explode, throw off, among other things, electrons, or particles of electricity. The discovery of this fact marks the starting point of the new physics, which teaches that electricity and matter are fundamentally the same. Electrons are thrown off by a hot metal also, but such electrons may be controlled and when captured in a vacuum tube, they enable us to produce radio broadcasting.

Radium and its salts produce three types of rays, called, respectively, alpha, beta, and gamma. Alpha particles are atoms of helium, each bearing two positive charges. They are projected from radium at a velocity that has been estimated at 9,000 to 12,000 miles a second. Their properties correspond to those of the canal rays in a vacuum tube; by deelectrification they pass into ordinary helium. Beta rays correspond to cathode rays of the vacuum tube. They appear to consist of negatively charged electrons or corpuscles projected from radium with a velocity that approaches that of light, though, since they are not homogenous, they have widely different velocities, up to a maximum of 170,000 miles a second. The beta particles are so much smaller than the alpha particles. however, that, although their average velocity is roughly ten times that of alpha particles, their kinetic energy is only about one-third as great. Gamma rays do not appear to be material particles at all; they are described as similar to, if not identical with, electromagnetic waves, or Roentgen rays, though they are penetrating and having a shorter wave length. Their velocity is given as 180,000 miles a second, a million times as fast as an express train traveling a mile a minute.

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[4] Other authorities place the period anywhere from 1,520 up to 2,500 years.

The three different types of rays may be separated by means of a powerful magnet. In an intense magnetic field alpha rays are repulsed from a magnet, beta rays are attracted to it, and gamma rays are not deflected either way. The penetration power of the different types of rays also varies in the order of 1:100:10,000 for alpha, beta, and gamma rays, respectively. This is demonstrated by the fact that although a few sheets of paper or a sheet of aluminum foil about 1 millimeter thick will cut off a large part of the radiations and although a sheet of lead about 5 millimeters thick will cut off nearly all of them, a residuum still remains unsuppressed even after passing through 15 centimeters of lead or a much thicker block of a less dense substance. The alpha particle rarely travels over 2 or 3 inches before it is stopped by impact with the oxygen and nitrogen molecules in the air; beta rays penetrate, on the average, about 2 feet in air and are always accompanied by gamma rays. With respect to ionizing power and in their effects upon an electroscope the three kinds of rays stand in reverse order, alpha rays being approximately 10,000 times as effective as the vastly more penetrating gamma rays.

The passage of gamma rays through matter is a complex phenomenon; it involves the production of secondary rays, but the precise manner in which it takes place is too difficult to elucidate. Much of the atomic energy of the radium atom is transformed into kinetic energy as the three types of rays are ejected, and its energy is soon transformed (upon recoil) into heat. The amount of this heat given off by 1 gram of radium has been estimated at 118 calories an hour,[2] or almost enough to raise 1 1/5 times its own weight of water from freezing to boiling temperature every 60 minutes.

## RADIUM EMANATION (RADON)

It should be noted that radiations from radium as ordinarily used are due largely not to the radium itself but to the radioactive products produced in it. Radium itself emits beta and gamma rays only feebly, and even its alpha ray activity is only about one-fifth as great as that it exhibits after it is about one month old. The penetrating beta and gamma rays arise almost entirely from the degeneration products of radium. Radium, like thorium, produces radioactive emanation which can be released from the radium by heating or dissolving it. This intensely radioactive "emanation" (or "radon" or "niton" as it is also called) has been widely used as a source of radiation in place of radium itself. The use of emanations possesses many advantages as a source of radiation. It has all the characteristics of a gas and can be readily transferred from one vessel to another. The radium itself can thus be used merely to produce emanations and is not disseminated or lost. The greater radioactivity of radon results in a small quantity having the same activity as a much larger quantity of radium; six millionths of a gram of radon (a curie) equals in gamma-ray activity a gram of radium.

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[2] Other authorities give 133 calories an hour.

The activity of the emanation is not permanent but decays to five-sixths in 24 hours to half value in 3.85 days and becomes very small (0.4%) after a month's interval. [6] Radium emanation, however, can be bubbled through a solution without losing its radioactivity.

The products of the decomposition of radium emanation are alpha particles and a radioactive solid, the atoms of which reform new and progressively more stable combinations. E. Rutherford found it convenient to analyze the active deposit of radium produced by the degeneration and decay of radium emanation into (1) the deposit of rapid change (radium A, B, and C); and (2) the deposit of slow change (radium D, E, and F). The manner of this degeneration will be further discussed in connection with the relation between uranium and radium.

## USES OF RADIUM

### General

The principal use of radium is for the treatment of cancer, but substantial amounts, estimated at as much as 10 per cent of the production in some years, are employed for the manufacture of luminous paints used on watch and clock dials, electric switch buttons, keyholes, and like products. War-time radium luminous material eliminated lights that would have betrayed the presence of troops to the enemy. Maurice Curie [7] has described experiments that indicate the profitable use of radioactive elements as fertilizers and for agricultural purposes. Tailings from Cornish ores were formerly shipped to France as fertilizers, but opinion in this country is that no good effect is obtained from radioactive substances as fertilizers.[8] Food-preservative receptacles have been made from mixtures of radiferous (carnotite) ore with white Portland cement, the idea being to prevent bacterial action through radioactivity. A few tons of carnotite are used annually in the manufacture of vessels designed to produce radioactive drinking water.

Radium is used to eliminate fire hazards in a large Russian rubber factory by preventing sparks of static electricity. The process is described as follows:[9]

> "When rubber is flowed upon a fabric base and dries upon it, enormous charges of static electricity are the result of the friction of the rubber-covered fabric against parts of the drying machinery. These accumulated charges finally result in a breakdown, and a discharge becomes inevitable. The spark resulting constitutes a great hazard, especially where highly explosive vapors are present. This is prevented when radium

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[6] Standard Chemical Co.'s figures.

[7] Curie, Maurice, Le Radium et les Radio-Elements. Encyclopedie Miniere et Metallurgique, Paris, 1925, ch. 7, 352 pp.

[8] Hess, Frank L. Mineral Resources of the United States, U. S. Bureau of Mines pt. 1, 1924, p. 471.

[9] Foote-Prints, Radium to Reduce Fire Risk: vol. 2, No. 1, 1929, pp. 28-29.

> is placed near the point where the electricity is generated, because the radium rays ionize the air and make it so good a conductor that dangerous charges flow harmlessly to the nearest metal part and thence to the earth. The cost is very low, as 1 milligram is sufficient to last for a few centuries at least."

The importance of radium in pure research can not be overestimated. Dr. Koller, 10/ of the research laboratory of the General Electric Co., has said:

> "The alpha particles shot off from radioactive atoms have been used as probes with which to search deep into the centers of other kinds of atoms. This method has been very fruitful and has given much information regarding the size and weight and atomic numbers of atomic nuclei. It has been possible by bombarding atoms with swift flying alpha particles to partially disintegrate them. Thus we have not only been able to observe the transmutation of the elements taking place in the radioactive series, but we have been able to cause it to take place at will in other substances, if only on a very small scale.

Paneth 11/ used radium emanation in testing the minute leakage of air through rubberized fabric for gas masks, and there are numerous scientific and technical problems in which radioactivity may be employed as a convenient indicator of conditions that the most refined and mechanical measurements fail to reveal.

In the laboratory radium has wrought a revolution in many of the preconceived notions of scientists. The existence of radium and of its decomposition products proves the transmutation of metals as an accomplished fact; and though the process of decomposition is far too slow to satisfy the commercial need for prompt returns, the fact that an atom, formerly considered an immutable and indivisible unit of matter, is actually a complex structure that may be altered to other atoms less complex goes far toward realizing the dream of alchemists. The study of the spontaneous disintegration of radioactive elements has given the scientific world an insight into the actual composition of matter; and radium rays, especially the alpha particles, have given physicists a new instrument with which to investigate the fundamental properties of material substance.

## Therapeutic Uses

Radium has long been used empirically for the treatment of cancer, but only in recent years have its biological and curative properties been studied systematically. That inexpert use was dangerous became known soon after radium was discovered, but suitable methods have now been elaborated for the treatment by radiu of cancer in almost every region of the body except the stomach. Many physicians are of the opinion that more and more radium surgery, will replace the use of the knife in the treatment of cancer.

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10/ Koller, L. R., The Radioactive Elements. Scientific Monthly, vol. 27, July, 1928, pp. 54-56.

11/ Paneth, F., Radio Elements as Indicators. 1928, p. 84.

Radium therapy is employed more or less experimentally for the treatment of sundry dermatological conditions and other diseases besides cancer, but medical interest is centered chiefly upon determining the best methods of application in the treatment of cancer. The influence of radium in various malignant conditions is marked, but the full extent of its field of usefulness for therapeutic purposes is not yet very clearly defined.

## Luminous Paints

Radium alone is not luminous. The faint glow that sometimes is exhibited by tubes of radium salts is owing to impurities. By mixing radioactive material with phosphorescent substances, notably with zinc sulphide, a paint that will glow in the dark may be produced.

According to Parsons,[12] it was a shoemaker of Bologna who noted over 300 years ago, that heavy spar heated in charcoal possessed the property of glowing in the dark after it had been exposed to light. Other phosphorescent powders mostly blends of zinc sulphide and alkali sulphide, were discovered subsequently, and were used for producing temporary stage effects, for example, long before the discovery of radium made it possible to maintain the phosphorescent glow for an indefinite period.

The ingredients and formulas for luminous paints undoubtedly change from time to time. In the United States the paint used on watch dials has consisted mainly of crystalline zinc sulphide (Sidot's blende), mixed with various proportions of radium, mesothorium, and radiothorium in order to obtain the desired amount of luminosity. Pure zinc sulphide may be prepared by repeated roastings and reprecipitations, and small amounts of cadmium, copper, and manganese may be added to give better luminescence. To an emulsion of the zinc sulphide mixture, radioactive material is added in the form of a soluble salt, which is then precipitated out as an insoluble sulphate. In the beginning the zinc sulphide was rendered luminous by activation by radium alone; but later, partly because they were cheaper and partly because they produced greater practical luminosity, mesothorium and radiothorium also were employed.

As reported by Doctor Martland,[13] the paint used by girls in a New Jersey factory contained chiefly zinc sulphide, rendered luminous by activation with about 20 to 30 per cent radium and from 70 to 80 per cent mesothorium containing radiothorium. He quotes other authorities to the effect that these paints may contain all the way from 0.7 to 3 and even 4 milligrams of radium element to 100 grams of zinc sulphide. Impurities may be added to the zinc sulphide as follows: Cadmium, 0.05 per cent; copper, 0.001 per cent; manganese, 0.0002 per cent.

British practice is described by Parsons.[14] Luminous paints are of military significance, being used not only for watch dials but for the illumination of gun sights and compass cards and, in general, wherever other forms of illumination at night might betray one's presence to the enemy. During the Great War, the British Government bought about 18 grams of radium for these purposes, and it is stated that at one period of the war the monthly consumption of radium for the manufacture of luminous paint amounted to 500 milligrams.

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[12] Parsons, A. T., Radium, with Special Reference to Luminous Paint. Jour. Oil and Colour Chem. Assoc., vol. 12, Jan., 1929, pp. 3-28.

[13] Martland, Harrison S., Occupational Poisoning in the Manufacture of Luminous Watch Dials. Monthly Labor Rev., U. S. Bureau of Labor Stat., vol. 28, June, 1929, p. 65.

[14] Parsons, A. T., Radium, with Special Reference to Luminous Paint. Jour. Oil and Colour Chem. Assoc. (British), vol. 12, Jan., 1929, p. 3.

## SUBSTITUTES

The Tariff Act of 1922 provided for "radioactive substitutes." these substitutes consist essentially of mesothorium which as previously noted may replace radium in luminous paints,. Viewed from a national or from a humanitarian standpoint, knife surgery, x-ray therapy, and certain kinds of medical treatment compete with radium in its theropeutic uses,

## BIOLOGICAL EFFECTS OF RADIUM EXPOSURE

Radium, though primarily useful in the treatment of diseased conditions of the human body, should be handled cautiously. All three types of rays from radium can produce intense local action on living tissues and, if allowed to act long enough and with sufficient intensity, will actually destroy living cells. Radium burns are serious and ordinarily do not develop for 10 to 15 days after exposure. If a tube containing, let us say, 20 milligrams of radium were held in the fingers for some minutes or if an unsealed tube of radium salt were brought near the skin for a short time, after a fortnight a soreness would be felt, which might develop eventually into a painful ulcer or even into cancer. The alpha and beta rays, though not so penetrating, are more destructive than the gamma rays; but the latter are the more insidious in their general effect on health, as they tend to produce, among other damages, pernicious anemia.15/ The effects of prolonged and incautious exposure have been demonstrated all too hideously in the sufferings of workers using luminous paint and the horrible deaths of several of the pioneer scientic investigators.

Continued exposure to radiation usually produces the following harmful effects: 16/

1. Pain, sensitiveness, or anesthesia of the skin of exposed fingers on hand.

2. Burns or destruction of the skin and underlying tissues.

3. Effect upon the blood and blood-making organs—usually a profound leucopenia affecting both the polynuclears and lamphocytes; a decrease in blood platelets; also a milder anemia accompanied by a high-color index. The reduction of the lymphocytes seems to be definite and regular; the effect upon the polynuclears is irregular.

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15/ Parons, A. T., Radium, with Special Reference to Luminous Paints: Jour, Oil and Colour Chem. Assoc. (British), vol. 12, Jan., 1929, p.7.

16/ Williams, R. C., Preliminary note on Observations Made on Physical Condition of Persons Engaged in Measuring Radium Preparations. Public Health Reports, Reprint No. 889, U. S. Public Health Service, Dec. 21, 1923, 24 pp.

4. Sterility.

5. The inhibition of the absorption of fat in the intestinal canal. This has been demonstrated experimentally.

The extent of the effect of the exposure to radiation depends upon:

1. Amount of radium to which the subject is exposed.

2. Length of time of the exposures.

3. Frequency of the exposures.

4. Proximity of the radium to which the subject is exposed.

5. Amount and character of protection afforded against radiation.

6. Character of the rays to which the subject is exposed.

The following is quoted from J. C. Mottram [17] regarding the biological action of radium:

> "Surveying the biological action of radiation, one of the first generalizations which may be made is that the various tissues differ widely in their susceptibility; some—for instance, nerve cells— show no changes after relatively large exposures, whereas others— for instance, reproduction cells—are altered by small amounts of radiation. The following tissues are especially sensitive; skin, blood vessels, connective tissue, hair follicles, reproduction cells, lymphoid tissues, and blood cells. Experimental evidence goes to show that the last three are more susceptible than the others, so much so that these tissues would be especially chosen for examination in searching for the earliest effects of radiation. There is no doubt about the sensitiveness of the reproduction cells. The sterility of X-ray workers who have good health in other respects is very clear evidence. As regards the blood changes, it may be mentioned that experiments on rats have shown that, by their blood changes, X-radiation could be detected where a photographic plate gives no record. The present state of our knowledge would lead to the conclusion that in the absence of blood changes the worker had received no more than a harmless amount of exposure to radiation."

An inspection of 31 factories by the United States Bureau of Labor Statistics revealed that a large number of workers were subjected to an exposure, possibly harmful) to radioactive substances. On the other hand, Swiss health inspectors subsequently made an investigation of conditions in Swiss watch factories using radium and reported that no cases of illness or harmful exposure were found. A the Monyco factory at La Chaux de Fonds, a nonwater varnish is applied in using the luminous materials, and steel or wooden pens are generally employed. Hair brushes are very rarely used, and every new workman is warned that it is dangerous to lick the brushes and is ordered to wash his hands carefully before leaving the room.[18]

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[17] Mottram, J. C., The Use of Blood Counts to Indicate the Efficiency of X-ray and Radium Protection. Brit. Med. Jour., vol. 2, 1921, p. 269.

## COLORING EFFECT OF RADIUM RAYS [19]

Radium salts and solutions color glass and rock crystal a deep purple, fluorspar blue, rock-salt yellow, and selenite brown. Kunzite becomes first colorless and then green. Even the diamond is colored by radium rays, and the green color that is produced by direct radiation either from high-grade salts or from radium emanation is not destroyed by sunlight, though it can be removed by heating the diamond to a dull-red heat for half an hour or less or by recutting (possibly as the result of the heat produced in the cutting process). The colors produced in other inorganic substances likewise may be discharged by heating. All kinds of glass fluoresce in radium emanation; certain kinds are colored purple, and others (containing selenium or cobalt oxide) take on brown shades.

## RADIUM MANIPULATION [20]

In carrying out manipulations with radium, the special problems to be considered arise from the extraordinarily high value of the material, the small quantities dealt with, and the danger to the health of the operator.

As a precaution against loss, apart from such obvious measures as carrying out all manipulations over large trays and dishes, the important thing is to make complete check measurements at every step and to throw away nothing until all the radium has been accounted for. It is useful to bear in mind the low solubility of radium sulphate and the useful circumstance that even the small amount of radium sulphate that does pass into solution is absorbed by precipitating barium sulphate in the liquid.

With regard to precautions, it is of fundamental importance to bear in mind (1) that radium itself gives only alpha rays, which are stopped by about 1½ inches (3.4 centimeters) of air or about 0.03 millimeters of water, and (2) that the first product of its disintegration is a gas. As long as radium is in solution and the emanation is allowed to escape (by means of a flue or draft), we have only alpha rays to deal with, and practically none of them can escape from the liquid. When the radium has been brought to a solid state, the greater part of the emanation remains in the salt, and the material begins to exhibit beta and gamma ray activity. Since this activity reaches half its maximum value in four days, three quarters in eight days, and so on, it folllows that operations on solid radium are most safely carried out as soon as possible after it has been brought into the solid state.

## HISTORY

The way to the discovery of radium was opened in 1895 by Roentgen, who found that the glow from a Crooke's tube contained penetrating rays, which he called

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[18] Buscher, Hugo, No.111. Effects after Working with Radium Twenty Years. The Manufac. Jeweler, vol. 86, No. 1, Jan. 2, 1930, p. 31

[19] Lind, S. C., and Bardwell, D.C., The Coloring and Thermophorescence Produced in Transparent Minerals and Gems by Radium Radiation. Am. Mineralogist, vol. 8, Oct., 1923, pp. 171-180, and Nov., 1923, pp. 201-208.

[20] Parsons, A. T., Radium with Special Reference to Luminous Paints. Jour. Oil and Colour Chem. Assoc. (British), vol. 12, Jan., 1929, pp. 14-15.

X-rays. Prof. Henri Bequerel, while investigating the effect of various phosphorescent substances, found that uranium salts produced photographic impressions even when enveloped with opaque substances. To Marie Sklovouski, a young Polish student, who later became Madame Curie, Professor Bacquerel delegated the task of learning how and why uranium possessed power to emit these peculiar rays, which he had proved to be electrical in character. Madame Curie, examining by electrical methods the radioactivity of a large number of minerals containing uranium and thorium, discovered that some specimens of pitchblende had about four times the activity of the metal uranium; that chalcolite, the crystallized phosphate of copper and uranium, was twice as active as uranium; and that autunite, a phosphate of calcium and uranium, was quite as active as the same weight of pure uranium. In order to check these discoveries, she prepared chalcolite artificially, starting with pure products, but found that this artificial chalcolite had only the activity represented by its composition, or, roughly, 40 per cent of the activity of uranium. This led to the conclusion that there was some element or substance in the residue from uranium minerals that possesses a high degree of activity. After an exhaustive chemical investigation of pitchblende from Joachimsthal, she found that this mineral contained not only uranium but also another radioactive substance, to which she gave the name of polonium, in honor of her native land. Later, in 1898, Monsieur and Madame Curie found still another element, which, when brought to a state of concentration, was several million times as active as uranium, and to this was given the name of radium. Debierne afterwards found a fifth radioactive substance, actinium; and in 1906 Boltwood isolated the metal ionium. Strictly pure radium chloride was first produced in 1902. Development in the knowledge of radioactivity has been rapid since that time.

Not long after the discovery of radium, the Curies brought it about that preparations of barium containing radium could be bought commercially through the Societe Centrale de Produits Chimiques. This company, under the direction of Debierne, was the first to undertake the treatment of pitchblende residues for the Curies. Another factory was soon started in Germany under the direction of Giesel, and within a short time Auer von Welsbach, Hittinger, and Ulrich were operating a plant in Austria.

For some time radium was produced only from the pitchblende ores of Austri in fact the Austrian Government presented Madame Curie, for her first working material, with a ton of uranium residues from the State mine of Joachimsthal, in Bohemia. As these ores were a Government monopoly, a world search was at once begun for other ores that might yield this material.

Occurrences of autunite were known in Portugal, and veins of pitchblende had been found previously in Cornwall in England and in Schneeberg and Johanngeorgenstadt in Saxony. Subsequently, pitchblende was found in the United States, in Mexico, and in the Indies, where it was exploited for a short time. The most important find was carnotite in the United States, deposits of which extend into a wild and mountainous tract through the States of Colorado and Utah. In 1911, Joseph M. Flannery, of Pittsburgh, Pa., undertook the exploitation of these deposits, organizing the Standard Chemical Co. In 1912, 1,200 tons of carnotite, containing 8 or 9 grams of radium, left the United States for France, where a radium extraction plant had been established to compete with the Austrian works. Plants were later established in the United States, which produced 10.5 grams of radium in 1913 and 22.4 grams in 1914. Among the new companies organized were the Schlessinger Radium Co., of Denver, Colo.; the Chemical Products Co., also of Denver; the Cummings

Chemical Co., of Lansdowne, Pa.; and the Radium Luminous Materials Corporation, of New York. The exportation of carnotite to Europe continued up to the outbreak of the World War in 1914. Previous to January 15, 1914, all the radium mines in the United States were privately owned, but after that date the American Government reserved the title to the new mines on public land. While prospectors had the right to exploit the mines that they discovered, the mineral had to be furnished to the American Government at a price fixed from time to time by the Department of the Interior. 21/

As early as 1912, the Bureau of Mines recognized the need of investigations to prevent waste of low-grade carnotite, to enable the miner to receive better prices for his product, and to insure a maximum profitable recovery. Dr. James Douglas, of New York City, and Dr. Howard A. Kelly, of Baltimore, Md., were interested, and before long the National Radium Institute was incorporated for the purpose of studying the best methods of producing uranium, vanadium, and radium, and the mining and concentration of ores from which these products might be obtained for use in radium therapy. A little later, at the suggestion of the institute, a cooperative agreement was made under which the Bureau of Mines undertook to provide the services of skilled chemists and mineral technologists to supervise the mining and concentrating of carnotite, the necessary funds to be furnished by the National Radium Institute. Under this agreement, a radium recovery plant was built in Denver, Colo., which began commercial operation in June, 1914. The aggeement between the National Radium Institute and the Bureau of Mines expired in 1916; but by the time the work ceased in January, 1917, about 8½ grams of radium had been extracted. The methods devised by the Bureau of Mines reduced the cost of recovering radium to about one-third of the then current price; and, in addition, methods of saving uranium and vanadium economically from these ores were perfected. By the expenditure of less than $38,000 of the Bureau's funds, the bureau obtained as its share of the results of the cooperative agreement more than $100,000 worth of radium. 22/

Although the domestic deposits of carnotite were relatively low grade, the United States maintained its lead in the radium industry for several years. The first discovery of pitchblende in the Belgian Congo was made in 1913 in the Luiswishi copper mine, and the second discovery of this mineral was made in 1915 in the Chinkolobwe (Kasalo) mine of the same company. Systematic prospecting begun in 1921 opened up other uranium-bearing veins. As this early work indicated the existence of a substantial supply of high-grade ore, a factory for the treatment of the Congo material was established at Oolen, near Antwerp. The first cargo of ore arrived at Antwerp on December 5, 1921; and under the direction of N. Leeman, assisted by M. Clerin, a suitable process of extraction was developed, so that the first unit of the new plant was in operation on July 8, 1922. Continued prospecting confirmed the existence at Chinkolobwe of considerable quantities of rich ore, yielding as much as 60 per cent of uranium oxide; and the works at Oolen were enlarged, until during 1925 they were supplying regularly nearly 4 grams of radium a month. A further increase in capacity was made in 1929.

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21/ Data in this paragraph are taken largely from the Smithsonian Report for 1925, The Manufacture of Radium, by Prof. Camille Matignon, Publication 2836, 1926, P.222.

22/ Kithil, Karl, and Davis, John A., Mining and Concentration of Carnotite Ores. Bull. 103, Bureau of Mines, 1917, p. 8.

The commercial history of the radium industry has been a series of monopolies—first Austria, then the United States, and finally Belgium having dominated the world situation.

## WHAT IS AN ATOM?

One of the most ancient of philosophies attempted to account for the universe and even life itself as constituted of simple, indivisible, and very tiny particles or atoms. About 460 B. C., Democritus, for example, encompassed in the theory the human soul, which, he asserted, consisted of atoms that were fine, round, and very smooth. 23/ As modern chemistry developed the fact that elements unite witl one another in definite combining proportions, the atom once more assumed importanc as the smallest portion into which matter can be divided and still preserve its identity. It became the chemist's unit of measurement, and the weights of atoms of various elements were compared numerically with the weight of the lightest known atom, a particle of hydrogen.

Only with the discovery of the radioactive elements came the knowledge that atoms themselves were composed of even smaller units, and this was not conclusively demonstrated until after Sir J. J. Thomson proved in 1897 the existence of electrons in the so-called cathode rays. Highly precise measurements of the mass and charge of the electron were later made by Prof. R. A. Millikan.

The atom has now been identified as a miniature solar system, made up of protons and electrons. Both of these componets are charges of electricity, the protons being positive and the electrons negative charges. At the center of each atomic universe is a small compact sun, which may be composed of both positive and negative charges, but with some excess of the former. Around this sun rotate planetary electrons. The exact number is determined by the net positive charge (that is, excess of protons) of the nucleus. Hydrogen, the simplest atom, has only one proton as a nucleus and consequently only one electron rotating about it. The atom next in size as far as is known, is helium; its nucleus comprises four protons and two electrons; and two more electrons rotating about this nucleus balance the two excess positive charges at the center. The nucleus of lithium, which comes third in the series, contains seven protons and four electrons; and the net positiv charge of these on the nucleus is balanced by three negative charges in the form of rotating electrons.

## RELATION OF RADIUM AND URANIUM

Uranium has been called the "mother of radium"; but (still retaining the metaphor) ionium is more strictly the mother and uranium may be more accurately described as a maternal ancestor. The successive steps in the transmutation of uraniu to radium and its further degeneration into radium lead are shown in the accompanyir table. Freshly purified uranium (as noted in the history of Madame Curie's work leading to the discovery of the new element) emits only alpha rays. But beta rays are soon developed (uranium X); and wherever uranium occurs in nature, it is accompanied by radium. These elements are far too uncommon for this association to be merely the result of chance; and, moreover, the proportions of radium to uranium in old uraniferous minerals is always the same, approximately 1 part in 3,400,000. This figure is evidently an equilibrium ratio, and uranium minerals do not always contain this maximum or theoretical quantity of radium. Some authorities have deemed

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23/ Standard Dictionary, p. 180.

that this condition was the result of a natural leaching out of radium, but the opinion of most scientists is that such minerals are not yet old enough for equilibrium to have become established.

Radium appears to have lost 3 atoms of helium (atomic weight 4) since its atomic weight is approximately 226, or 12 less than that of uranium. Beta and gamma rays are also lost, but these evidently have no measurable weight. As already noted, an atom of radium itself loses an atom of helium (with a double positive charge) and becomes the new gaseous element, radon or radium emanation. This change is very slow, about one-millionth of the radium atoms changing in a day; so that it is only after 1,690 years of such change that even half of the atoms in a given mass of radium will have decomposed. Radon, however, decomposes much more rapidly than does radium -- half of any quantity of radon transforming in 3.85 days, half of the remainder in the next 3.85 days, and so on.[24] Radium A, the next product, is quickly decomposed, its half life being but three minutes. It likewise emits a helium atom (alpha ray); but radium B, which lasts only a few more minutes, emits only beta and gamma rays and does not measurably lose in atomic weight as it changes into radium C. As radium C decomposes, there comes a split in the family tree. Most of the atoms (99.97 per cent) are transformed into radium C', emitting beta and gamma rays; but a small part (0.03 per cent) is changed into radium C", emitting alpha rays. Radium C" almost instently is transmuted into radium D and eventually becomes radium lead. Radium F, or polonium, one of the intermediate products, finds a place in the periodic table as an element similar in many respects to tellurium, which falls in the same column of the periodic table.

Uranium-radium series

| Element | Radiation emitted | Time to deacay to half value | Atomic weight | Average life |
|---|---|---|---|---|
| Uranium I | Alpha | 5,000,000,000 years<br>(4,600,000,000 years) | 238.2 | 8,000,000,000 |
| Uranium X | Beta | 24.6 days | 234 | 35.5 |
| Uranium X2 | Beta and gamma | 1.15 minutes | 234 | ? |
| Uranium II | Alpha | 2,000,000 years | 234 | 2,000,000? |
| Ionium | Alpha | 69,000 years | 230 | 200,000 yea |
| Radium | Alpha | 1,690 years | 226 | 2,500 years |
| Radon (emanation) | Alpha | 3.85 days | 222 | 5.6 days |
| Radium A | Alpha | 3.0 minutes | 218 | 4.3 minutes |
| Radium B | Beta and gamma | 26.8 minutes | 214 | 38.5 " |
| Radium C | (Alpha(99.97%)<br>(Beta and gamma(0.03%) | 19.5 minutes | 214 | 28.1 " |
| Radium C' (99.97%) | Alpha | 0.000001 seconds | 214 | |
| Radium C" (0.03%) | Beta | 1.4 minutes | 214 | |
| Radium D | Beta (soft) | 16.5 years | 210 | 24 years |
| Radium E | Beta (soft) | 5.0 days | 210 | 7.25 days |
| Radium F (polonium) | Alpha | 136 days | 210 | 202 days |
| Radium G (Z') (lead ?) | None | Stable | 206 | |

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[24] Robinson, G. A., and Miller, A. L., Radon and its Therapeutic Uses, Address before Rensselaer County Medical Soc., Troy, N.Y., Oct. 2, 1928, printed by the Standard Chemical Co., New York.

## ACTINIUM SERIES

Uranium when it disintegrates apparently does not all follow down the series of changes that includes radium. Uranium ores contain actinium, but in much smaller quantities even than radium. According to some authorities, uranium when it decomposes gives birth to both families, 92 per cent going to form the radium series and 8 per cent proceeding through the much shorter series of transformations that constitute the actinium series. As in the case of the radium family, the end product is lead. Actinium is extracted from pitchblende; it is remarkably similar in many of its properties to lanthanum.

The actinium series is of less economic significance than the radium or mesothorium, partly because of the difficulty of separating it in quantity from the rare earths with which it occurs, but more especially because of its short radioactive life. There is only one gaseous product, or "emanation," and this emanation emits only alpha rays, and these only briefly.

Actinium series 1/

| Element | Radiation emitted | Average life | Possible atomic weight |
|---|---|---|---|
| Uranium Y ................. | Beta | ? | 234 |
| Protoactinium ............... (uranium Z, eka-tantalum) .. | Alpha | ? | 234 |
| Actinium .................. | Alpha | ? | 230 |
| Radioactinium ............. | Alpha | 28.1 days | 226 |
| Actinium X ................ | Alpha | 15 days | 222 |
| Emanation ................. | Alpha | 5.6 seconds | 218 |
| Actinium A ................ | Alpha | 0.003 seconds | 214 |
| Actinium B ................ | Beta | 52.1 minutes | 210 |
| Actinium C ................ | Alpha | 3.1 minutes | 210 |
| Actinium D ................ | Beta and gamma | 6.83 minutes | 206 |
| Actinium E ................ (lead) | None | unknown (stable ?) | 206 |

## RADIO ELEMENTS AND THE PERIODIC CHART OF MENDELEEF--ISOTOPES

Despite the revolutionary nature of the discovery of radioactivity, the new elements of the three radioactive series are all fitted readily into the familiar periodic arrangement credited to Mendeleef. This link with the older chemistry contributes to a clearer conception of the nature of the radioactive elements and the mechanics of their transmutation. A simple explanation of the progressive transmutation of uranium into radium and eventually into lead is that uranium, the heaviest of all known elements, gives off helium, 1 atom at a time, and suffers a loss of 4 points in atomic weight as each atom escapes. In forming radium, 3 atoms escape; and by the time lead alone remains, 5 more (a total of 8) have been dissipated in the form of alpha rays. Radium falls in place in the column of the alkaline--earth metals, where it properly belongs by virture of its proper-

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1/ Adapted from article by Soddy, F., and Cranston, John A., The Parent of Actinium. Proc. Royal Soc., vol. 94 A, London, 1918, pp. 384-404.

ties, valence, and atomic weight; radon (emanation) similarly finds an appropriate position in the column of inert gases, having a valence of zero; and actinium in the column of trivalent elements occupies a position that was recognized as representing an unknown element even before its existence was otherwise suspected.

For many of the new elements, however, no gaps in the periodic table existed, and numerous tests have established the chemical inseparability of several elements of the radioactive series, such as thorium and radioactinium or radium and mesothorium. In order to explain the conditions whereby two or more substances occupy the same place in the periodic table, it became necessary to create the concept of "isotopes." Isotopes, according to Hendrick,[25] "are atoms that have the same atomic number—that is, the same number of free positive charges within the necleus and opposed electrons outside it, but not the same number of hydrogen or helium atoms packed about the nucleus. Therefore, the atomic weights of isotopes differ. And while the weight of each atom is integral, mixtures of isotopes gige fractional weights." The theory of isotopes has received direct verification as a result of the identification of several kinds of "lead" resulting from the degeneration of radioactive substances.

## GEOLOGICAL OCCURRENCE

Radium is widely distributed in the earth's crust, but generally in infinitesimal quantities. Almost all rock samples, when examined by the most sensitive methods, show an extremely weak radioactivity. According to measurements made by Strutt, the terrestrial crust contains, on the average, about $4 \times 10^{-6}$ grams of radium per cubic meter.[26] Igneous rocks, notably granites, are most active; but only the pure limestones and quartz sands are nearly inactive. The waters of various mineral springs that have been in contact with radioactive earth are almost always more or less radioactive.

Radium minerals are generally found in connection with granite masses, that is, where granite forms at least part of the rocks of the country. When original minerals, notably pitchblende, break down through weathering, other minerals are formed, such as autunite, torbernite, carnotite, and tyuyamunite.

## RADIOACTIVE MINERALS

The ores of radium are uranium ores. They are of three general types: (1) uraninites, (2)-niobium-titanium tantalates of the rare earths and uranium, and (3) secondary minerals.

Type 1.- The uraninites, which are the richest in uranium, include such crystalline varieties as broeggerite, cleveite, and certain uraninites found in the United States and East Africa. These varieties occur in pegmatites in well-developed crystals, though these crystals rarely exceed 2 centimeters in diameter. Pitchblendes are amorphous uraninites, and they occur in metalliferous veins, especially associated with sulphides of silver, lead, nickel, zinc, and other metals. The complex primary minerals of the second group, which are best represented by the betafite deposits of Madagascar, are often well crystallized and occur in pegmatite veins. Euxenite, however, is more often found in compact masses. The various secondary minerals include phosphates, carbonates, arsenates, sulphates, silicates, vanadates, and uranates. Most of them are characteristically colored bright green or yellow. The most important are autunite and carnotite. Being formed relatively recently, many of these secondary minerals have not yet had an opportunity to reach equilibrium. The uranium-radium ratio, therefore, may vary over fairly wide limits.

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[25] Hendrick, Ellwood, Atoms and Isotopes. Chem. and Met. Eng., vol. 26, Mar. 29, 1922, p. 586.

[26] Matignon, Camille the Manufacture of Radium. Smithsonian Rept. for 1925, Publication 2836, 1926, p. 224.

The terms "uraninite" and "pitchblende" are often used synonomously, but the term "pitchblende" is applicable strictly only to the impure amorphous form of the mineral, often with botryoidal surfaces and a conchoidal fracture. Uraninite, when crystallized, belongs to the isometric system. It is black or grayish-black in color, often with a glossy or pitchlike luster. The hardness is approximately that of steel (5.5), and it is heavy. Its specific gravity, however, ranges from 5 to 9.7 according to its purity. The formula is somewhat indefinite, and it is usually described as a uranate of $UO_2$, together with variable quantities of lead, calcium, iron, copper, bismuth, thorium, zirconium, and other rare and common elements.

Type 2.- Carnotite is an amorphous, soft, powdery material, occasionally talcose or waxy in character. The color is generally brilliant canary yellow, though it may be discolored by iron, organic matter, or other substances. It is essentially a hydrous potassium-uranium vanadate and is sometimes associated with the hydrous calcium-uranium vanadate, known as tyuyamunite. It is the principal ore mineral in the United States. Autunite, found essentially in Portugal, is a phosphate of uranium and lime, containing as much as 50 per cent of uranium. The color is described as greenish-brown to sulphur-yellow. It is soft (hardness 2 to 2½) and rather light (specific gravity 3.5 to 3.9). Torbernite is a mineral of similar composition, containing copper instead of calcium, and it is green in color.

Type 3.- In addition to betafite, the niobium-titanium tantalates include, samiresite, ampangabeite, euxenite, blomstrandite, samarskite, fergusonite, and other minerals. These mineral substances all have common characteristics and are easily recognized because of their high density (over 4), black or dark-brown color, and greasy, brilliant fracture. Minerals of this group were first discovered in Norway and Greenland (euxenite, blomstrandite, samarskite, and fergusonite). Later a deposit of euxenite was found in Brazil, but now by far the most important deposits are those in Madagascar, which furnish several unique varieties, notably betafite, samiresite, and ampangabeite, all of which are rich in uranium. Certain minerals of this group, however, contain thorium in larger or smaller amounts, and as a result the radium extracted from them may contain substantial quantities of mesothorium.

The principal thorium mineral is monazite, an anhydrous phosphate of the rare earths cerium, lanthanum, neodymium, praseodymium, ytrium, and erbium. Thoria is not an essential constituent but is always present in quantities ranging from 1 per cent to 12 per cent. Monazite yields a little radium, as well as mesothorium. A ton of monazite containing 5 per cent of thorium will contain about 6.4 milligrams of mesothorium element. In addition, it contains a little radium, and the presence of uranium has also been established.

There are probably 100 or more minerals known to contain uranium. A list including all the more important minerals will be found in Appendix I.

## IDENTIFICATION

Since radioactive minerals accelerate the rate of discharge of static electricity from a charged body, an electroscope is commonly used as a means of detecting the presence of radium or uranium in ores. A common form of electroscope consists essentially of a strip of metal foil (silver or aluminum) suspended at the top in such a manner as normally to be in contact with a vertical rod of metal. When both leaf and rod are charged with electricity, however, the leaf is repelled from the rod and stands away from it at a more or less wide angle. The whole device is inclosed in a compartment with mica windows to exclude air currents and other disturbing conditions. But in every instrument there is some leakage; and as the intensity of the charge diminishes, the leaf drops back slowly to its normal position. When a radioactive substance is introduced into the machine, the rate of discharge is greatly accelerated.

The electroscope can be used even for quantitative determinations, since the rate of fall is, within reasonable limits, always proportional to the amount of uranium oxide (or radium) present. The fall can be observed through a telescope equipped with a graduated scale, and the rate of fall can be measured by means of a stop watch in terms of the divisions on the scale. By comparing the rate of fall with an unknown sample against the rates from standard ore of known uranium content and correcting for the natural leak, the percentage of uranium oxide in the unknown sample may be estimated according to the formula: (Rate of fall with sample minus rate of natural leak) : (Rate of fall with standard minus rate of natural leak) :: (Per cent $U_5O_8$ in sample) : (Per cent U O in standard).27/

According to Hess: 28/

> "A test for radioactivity is not a conclusive test for radium and uranium, for thorium also has a disintegration product, mesothorium, which closely resembles radium in its behavior. Tests for radioactivity will therefore show only whether the specimen contains radium and uranium or mesothorium and thorium or all four. The appearance of the minerals will generally determine which of the elements is present, but in case of doubt the determination requires more elaborate tests, which should be made by an experienced chemist.
>
> "To test for radioactivity, lay the mineral to be tested on a plate holder containing a sensitive photographic plate; and if uranium is present in any considerable quantity, the plate will be "light-struck" within a period ranging from 12 hours to a week, the time depending on the quantity of radioactive matter in the specimen."

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27/ Kithil, Karl L., and Davis, John A., Mining and Concentration of Carnotite Ores. Bull. 103, Bureau of Mines, 1917, p. 45.

28/ Hess, F. L., Rare Metals in 1929. Mineral Res. of the United States, 1927, Bureau of Mines, 1930, pt. 1, p. 475.

"When objects are laid directly upon a sensitized film or plate, deceptive effects are obtained by pressure. Many supposed "radiographs" made by mere pressure have been received. In some of these the sensitized coating has been entirely removed under the object, and the remainder was so light-struck that the experimenter thought he had obtained a "reversed" effect."

## PROSPECTING

Since all uranium ores are "spotty" and irregularly distributed in scattered and usually relatively small deposits, prospecting constitutes in all localities an important factor even after commercial development has commenced. Many of the uranium minerals are brightly colored (either yellow or green), and even the black varieties have a characteristic appearance,' so that ore bodies are readily recognized if they outcrop. More often, however, they do not coutcrop, and then the search for ore involves a careful study of the geological conditions. In vein deposits, such as those in Cornwall or Czechoslovakia, the problem of finding ore may be somewhat simpler than it is in the patchy deposits of carnotite in the United States, but even in vein deposits there are ordinarily local differentiations in the appearance or the associations of the minerals in the vein filling that properly interpreted, may lead to the discovery of ore shoots with a minimum of "dead work" underground.

Kithil and Davis [29]/have noted certain indications for carnotite, but add that no rule in prospecting can be given, except that good judgment might be shown in choosing a favorable location. The erratic nature of the occurrences is indicate by the fact that an average of 14 per cent of the total time spent on working the claims operated by the Bureau of Mines in 1924 and 1925 was spent in searching for ore.

In prospecting and in delimiting ore bodies, after they are discovered, all the usual methods may be employed—test pits, tunnels, core drills, and churn drills. Even core drilling, however, is uncertain unless the holes are closely spaced, for there is usually little continuity to the deposits.

## MINING AND PREPARATION OF THE ORE

The patchy nature of the ore deposits precludes the adoption of modern, large-scale methods for the mining of radium-uranium ores. Gophering seems to be the only economical means for extracting the ore-bearing material, and in all districts hand-picking plays a leading role in preparing the ore for shipment.

Since most of the uranium minerals are relatively brittle, they tend to form an enriched dust when the ore is crushed. In Cornwall the mechanical treatment of the ore has consisted simply of drying, crushing, and screening on a fine screen; but by treating low-grade dump material (reject from hand-picking), concentrates containing from 9 to 10 milligrams per ton were obtained, or more than twice the averag content of the uncreened material. Table concentration was attempted in Australia but did not prove satisfactory. The Bureau of Mines did considerable work on concentration; the results of this work are given in Bulletin 103, pp. 61-76, by Kithil and Davis, previously referred to. Successful tests were made of a wet method. By using carnotite ore containing only 0.75 per cent uranium oxide ($U_3O_8$), it was possible to obtain concentrates containing 2.25 per cent by simply running the ground ore (80 or 100 mesh) through a Dorr classifier and saving the slimes; and doubtless much higher-grade concentrates and a satisfactory recovery could be made by elutriating the material, agitating it so as to liberate the more tenaciously adhering carnotite from the silica grains, and carefully controlling the subsequent settling operations.

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[29]/ Kithil, Karl L., and Davis, John A., Work cited.

Owing to shortage of water, however, dry methods were actually employed at the bureau's plant. The equipment included: a 5 by 9 inch Sampson crusher, a drier and fan, a cyclone dust collector, two sets of coils, two bucket elevators, an ore-belt conveyor, a tailings-belt conveyor, a vibrating screen, a Raymond No. 00 pulverizing machine, a tabular dust collector, several fans and cyclones, and a bag dust collector. The milling ore had an average content of 0.85 per cent uranium oxide ($U_3O_8$); the average uranium-oxide contents of the four kinds of concentrates were as follows: No. 1, 2.91 per cent; No. 2, 3.55 per cent; No. 3, 2.76 per cent; No. 4, 3.44 per cent. The total average of all four concentrates combined was 2.92 per cent uranium oxide. Almost 88 per cent of the total quantity of concentrates was collected as No. 1. The tailings averaged 0.37 per cent uranium oxide and at times as much as 63.7% of carnotite contained in the milling ore was extracted.

With respect to carnotite, the concentrating problem has been recently reviewed by Doerner. 30/

> "Carnotite ore is a sandstone that has been impregnated with carnotite and related minerals. Organic matter such as the remains of trees often caused deposition of "bug holes" of nearly pure mineral, but as a rule the carnotite forms incrustations on the sand grains on exposed faces, in joints and fractures of the rock, and less abundantly as individual grains. As the carnotite is relatively soft, most ores are easily crushed to the size of the sand grains.
>
> "These ores are concentrated by a mechanical separation of the dust from the coarser sand grains. This may be accomplished either wet or dry. Wet methods are more effective, but scarcity of water in the carnotite areas makes the use of wet methods impractical in most cases. By the dry method the dry, crushed ore is agitated in a current of air to separate the dust from the sand. For good results the sand grains must be scoured clean from adhering mineral, with the minimum crushing of the barren grains. Ordinary crushing or grinding equipment is not well adapted to this operation, because it is designed to crush or pulverize but not for scouring through attrition. The impact or beater mill used in the Bureau of Mines work 31/ is probably the best standard type for this purpose.
>
> "Early in 1921 the author completed semicommercial tests on the dry concentration of low-grade carbonaceous ores from the Temple Mountain area near Green River, Utah. Because of the large amount of organic material present, these ores are among the most difficult to treat by either mechanical or chemical methods. Much of the organic substance resembles a tough asphalt and is so closely associated with the valuable minerals that both

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30/ Doerner, H.A., Possibilities of Production of Radium and Vanadium from Carnotite. Ind. and Eng. Chem., vol. 22, No. 2, Feb., 1930, pp. 185-9. 31/ Kithil, Karl L., and Davis John A., Work citied.

mechanical and chemical disintegration are obstructed. It was found that the difficulty could be overcome by roasting, a procedure which was later patented by another worker.[32] Roasting not only removes all organic matter and moisture, but it also causes chemical changes which disintegrate the physical structure of the valuable minerals, reducing their powers of cohesion and adhesion so that they are more easily reduced to a dust.

"A special machine was designed and constructed to scour the sand and separate the dust. It consisted of a long metal trough provided with a tight hood and had a double row of agitating paddles. The latter were mounted on parallel, horizontal shafts revolving in opposite directions. The roasted ore was fed continuously into one end of the machine through a sealed hopper and subjected to the action of the paddles which tossed it about. Jets of compressed air in the bottom of the trough blew off the dust which was collected in the usual arrangement of cyclone dust collector and bags. The clean scoured sand was discharged through a sealed overflow at the end of the trough opposite the feed hopper. This equipment and procedure gave much better results than had been previously attained."

## EXTRACTION OF RADIUM SALTS

Because of the small quantities of radium present in its ores the economical extraction of radium presents problems substantially different from those encountered in other fields of metallurgy. It is not unusual to treat 200 tons or more of ore in order to recover 1 gram of radium, a yield of only one part in one hundred and eighty million. The high unit value of the material likewise introduces a new factor calling for an extraordinary amount of analytical and other technical control work. Many of the leaching and washing operations are tedious, and the time-consuming nature of the various steps in the different processes is reflected in the initial cost and relatively large overhead and carrying charges of a radium plant as compared to the volume of material that can be put through it.

All radium reduction processes known to the writer involve at some stage the precipitation of a radium-barium sulphate, which precipitate is converted into a soluble salt and purified by a long series of repeated fractional crystallizations. The National Radium Institute employed Nitric acid initially to break up the ore; and in the treatment of autunite from Portugal, hydrochloric acid is used. Fusion with sodium sulphate was the means for extracting radium from pitchblende long before the discovery of radium. The treatment residues which contained the radium were formerly discarded, but subsequently were treated first with caustic soda and then with acid (HCl), and finally with sodium carbonate, in order to extract the impurities and to convert the radium sulphate into radium carbonate. After washing free from sulphates the residue is treated with pure hydrochloric acid to dissolve the radium. Boiling with sodium carbonate and acid treatment has to be repeated three times, and this phase of the process alone consumes from four to six weeks.

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[32] Vogt, L. F., Concentrating Carnotite Ores. U. S. Patent 1,429,550. September 10, 1922.

The details of many processes have been adequately covered by other Bureau of Mines publications.33/ All important methods methods for extracting radium from carnotite were tested on a great variety of ores and critically compared by H. H. Barker34/ whose results have been summarized by Doerner,35/ as follows:

> "(1) Direct solution in an acid. - Nitric acid gives a higher extraction of radium from many ores than can be obtained with any other agent. Nitric acid is not efficient in extracting vanadium, and with some types of ore, notably the carbonaceous ore from Temple Mountain, its recovery of radium is also poor. The use of hydrochloric acid has not found general application, because it will extract a high percentage of radium from only a few ores. It is an effective solvent of vanadium, however. Hot, concentrated sulphuric acid is an effective solvent for both radium and vanadium, but its use involves mechanical difficulties which have not been satisfactorily overcome.
>
> "(2) Conversion to carbonate or oxalate, and solution in hydrochloric acid. - To convert the ore to carbonates or oxalates, it is either treated in an autoclave with sodium carbonate solution or simply boiled in an oxalic acid solution. After filtration and washing, the residue is treated with hydrochloric acid solution and again filtered. Radium, vanadium, and uranium are precipitated from the filtrates. These methods are fairly effective with most ores.
>
> "(3) Sulphating, sliming, and re-treatment of slimes.- The method involving sulphating, sliming, and re-treating of slimes is complex and has many variations. The ore is sulphated by heating with concentrated or dilute acid or by fusion with nitre cake. The slimes are then washed away from the sands and dewatered, and uranium and vanadium are recovered from the solution. The radium is recovered from the slimes by using method (2) or by repeated fusion with caustic soda, each fusion being followed by water and acid leaches. The latter variation is more expensive, but results in a higher recovery of radium."

Since the gangue material ordinarily consists largely of silica, the cost by fusing with an excess of caustic soda, or soda ash, and then leaching out the sodium silicate with water is prohibitive in the case of any but high-grade ore or concentrated products. The use of hydrofluoric acid was patented by McCoy.36/ More recently Fleck and Haldane37/have patented the use of a soluble fluoride ($NH_4HF_2$) and sulphuric acid; provision is made for regenerating the fluoride but at the cost of a considerable consumption of sulphuric acid and lime.

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33/ Moore, Richard B., and Kithil, Karl L., A Preliminary Report on Uranium, Radium and Vanadium. Bull. 70, Bureau of Mines, 1913, 114 pp.
Doerner, H. A., Notes on Extraction and Recovery of Radium, Vanadium, and Uranium from Carnotite. Rept. of Investigations 2873, Bureau of Mines, May, 1928, 12 pp.

34/ Barker, H. H., Experiments in the Extraction and Recovery of Radium From Typical American Carnotite Ores. Univ. of Missouri Bull.,vol. 24, No. 26, Series 23, Sept., 1923, 87 pp.

35/ Doerner, H. A., Page 3 of Work cited.

36/ McCoy, H.N., Production of Concentrated Radium Residues and the Separation of Radium Compounds Therefrom, U. S. Patent 1,154,230, 1915. 37/ Fleck, H., and Haldane, W. G., Art of Extracting Metallic Values from Ores, Concentrates, and the Like. U. S. Patent 1,577,217, 1926.

According to Doerner [38] it is not difficult to obtain a good recovery of uranium and vanadium as by-products from the treatment of carnotite by any of the usual methods of extracting radium, although at the present time no attempt is made to recover the radium even as a by-product from the production of vanadium. The usual, and probably the best, method of recovering vanadium from vanadiferous sandstone (roscoelite and carnotite) is to roast the ore with about 10 per cent of common salt and 2.5 per cent of soda ash. The calcine is first leached with water and then with dilute sulphuric acid, and vanadic acid, or iron vanadate, is precipitated from the leach liquors. This leaves any radium present in the ore as a fine precipitate in the tailings, and when these tailings are discarded the radium is lost beyond all hope of recovery. It would be practicable, however, to deslime the tailings before rejecting them, and to store the radium in the form of dewatered slime. Further reference will be made to this procedure in the section on cost of production.

## WORLD PRODUCTION

Altogether the world has produced only 550 to 600 grams of radium, or about 1¼ pounds. New production based upon most recent available figures is at the rate of less than 50 grams — say 1¾ ounces — a year. Since a considerable amount of radium has been used in luminous paint (particularly during the war) the world's stock of radium is doubtless substantially less than 300 grams. Even the value of these small quantities of radium scarcely measures the importance of the industry. On the basis of an average price of $90,000 per gram, probably a close approximation, the sales value of the radium produced since 1898 is not greatly in excess of $50,000,000.

No accurate statement can be made as to the world's total production of radium from the time of its discovery until the present time. From data (partly confidential) available at the Bureau of Mines, Hess [39] prepared estimates of the output previous to 1926; and subsequent comment on this estimate has tended to confirm it as the best approximation that can be made, except perhaps as regards the Cornish figures, which have consequently been slightly increased. The following table represents the estimated distribution of total production, according to sources of ore, up to December 31, 1928.

Total production of radium element by countries, 1898 - 1928

| Sources of ore | Grams of Radium element |
|---|---|
| United States | 250 |
| Belgian Congo | 245 |
| Czechoslovakia[1] | 45 |
| Portugal | 15 |
| Madagascar | 8 |
| Russia | 6 |
| Cornwall | 5 |
| South Australia | 1 |
| Total | 575 |

[1] Formerly Austria

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[38] Doerner, N.A., Possibilities of Production of Radium and Vanadium From Carnotite. Ind. and Eng. Chem., vol. 22, No. 2, Feb., 1930, pp. 185-189.

[39] Hess, F. L., Rare Metals in 1926. Mineral Resources of the United States, 1926, Bureau of Mines, 1929, pt. 1, p. 268.

In addition to the countries listed in the foregoing table, radioactive minerals have been reported in commercial amounts in Tonkin (Indo-China) which produced a few tons of autunite in 1911, in British Africa and, recently, in Canada.

Belgium is important as the country in which the Congo ore is reduced, and France has been prominent in the radium industry not only because of the work of the Curies in Paris but also because of its radium factories, which have treated ores exported from Portugal, Cornwall, Australia, and elsewhere. Germany, though active in almost all other branches of chemical industry, has never been an important factor in radium supply. At Annaberg, on the Saxony side of the Erzgebirge, silver-cobalt veins like those at Jachymov occur. At Johanngeorgenstadt, to the southwest, the veins contain tin and silver-cobalt ores and (where dolomite spar appears) they also contain pitchblende, as at Annaberg; in the Gottessegen mine, pitchblende occurs sometimes in masses 2 or 3 inches in diameter. These German mines do not appear to have produced any substantial amount of uranium or radium minerals, although they have been worked extensively for bismuth (ocher) and also for cobalt and nickel. Occurrences of radium have been reported in Brazil and also in Sweden. In the Tanganyika Territory (British mandate) radioactive minerals have been found associated with muscovite in the pegmatites of the Uluguru Mountains and Lukwangula Hills; masses of pitchblende weighing as much as 70 pounds are said to have been found.[40] In Japan, sandstone nodules impregnated with manganese and a little iron have been described as radioative.[41] A few tons of ore containing pitchblende have been shipped experimentally from Mexico, where it has been identified in a number of gold mines, (notably on the Terrazas estate, about 90 miles west and little south of Chihuahua). In India some uraninite in masses weighing as much as 36 pounds is found in mica mines (pegmatite) near Pichli and Banekhap in the Gaya district. Most of these occurrences, however, are only of mineralogical interest.

## DOMESTIC PRODUCTION

Since 1923 very little radium has been isolated in the United States, and only a small quantity of radium ore has been mined. Almost one-half of the world's stock of radium, however, has come from American mines, and for about 10 years (1913-22) this country dominatod the world situation. This condition was changed abruptly when Belgium dropped the price of radium in 1923, but the stoppage of mining in the United States was the result solely of changes in economic conditions. Domestic deposits are by no means exhausted and under suitable conditions might be depended upon to yield a fairly large additional quantity of radium.

### Carnotite Deposits

Carnotite mining began in Colorado in a small way even before the discovery of radium. Parsons[42] records that as far back as 1881 Andrew J. Talbert mined some of the ore. This ore, however, was said to contain $5 a ton in gold, whereas most carnotite contains no precious metal values. About 1896, Kimball and

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[40] Committe of Civil Research, Radium Subcommittee Report (Cmd. 3313) London, 1929, p. 16.

[41] Limori, S., Manganiferous Radioactive Nodules from Tanokami, Province of Oomi. Sci. Papers Inst. Phys.Chem.Res., Japan, vol. 4, 1926, pp. 79-83.

[42] Parsons, Chas. L., Our Radium Resources. Jour. Ind. Eng. Chem., vol. 5, No. 11, Nov., 1913.

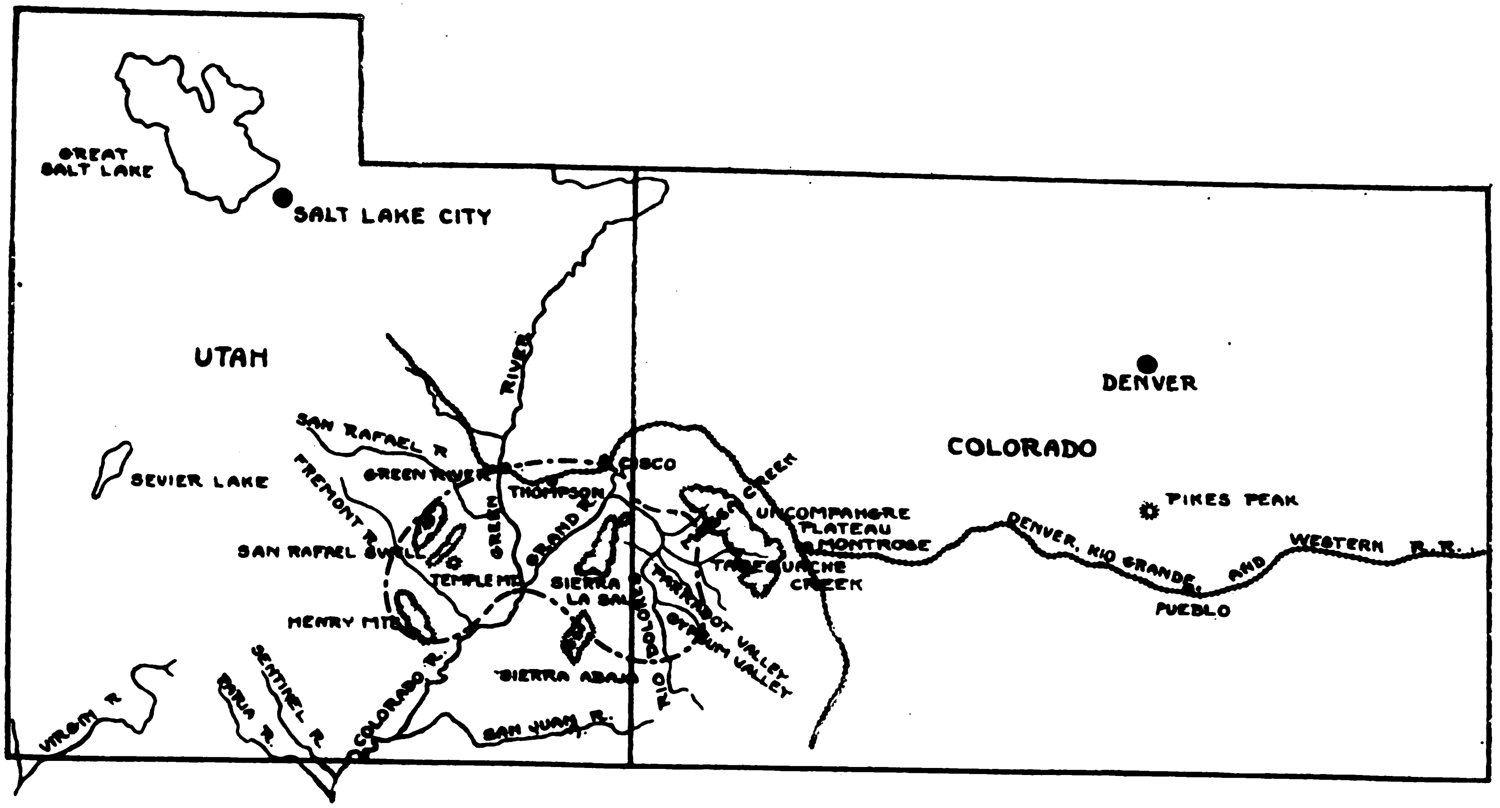

Figure 1.- Map showing main carnotite producing area in United States

Logan, after being informed by the Smithsonian Institution that their specimen contained uranium, mined 10 tons of ore and sold it in Denver for $2,700. In 1899 Poulot and Voillegue sent specimens to France and Fieldel and Cumauge named the mineral "carnotite" in honor of M. Carnot, then president of the French Republic. During the next three years Poulot and Voillegue produced 15,000 pounds of uranium oxide in the Paradox Valley. In 1903 their mill was started up again by the Western Refining Co., but it ran only one year. Shortly thereafter a new mill was built near it by the Dolores Refining Co., but after running for a few years it, too, was shut down. In 1911 Joseph M. Flannery, of Pittsburgh, Pa., organized the Standard Chemical Co., to exploit carnotite deposits as a source of radium. The pioneer work of the Bureau of Mines during the next few years has already been described (under "History"). Among the private companies subsequently organized were the Schlesinger Radium Co., of Denver, Colo.; the Chemical Products Co., also of Denver; the Cummings Chemical Co., of Lansdowne, Pa.; and the Radium Luminous Materials Corporation, of New York. In 1921, when domestic production was at its height, radium in salts was produced by the Standard Chemical Co., Pittsburgh; the Radium Co., of Colo., Denver; the United State Corporation, New York,; the W. L. Cummings Chemical Co., Philadelphia; and the Keystone Metals Reduction Co., Cheswick, Pa., In 1923 the Standard Chemical Co., the largest American producer, treated only accumulated ore on hand in its reduction plant, and assumed the agency for the sale of Belgian radium in the United States. The situation of other companies in that critical year was thus described by Hess:[43]

> "The United States Radium Company was the only American firm that isolated radium in 1923 from currently mined ores. * * * Although the Radium Company of Colorado, at the time the Katanga product threatened the market, had just developed at Camp Maverick, near Gateway, Colorado, the best bodies of ore in its history, the company found it necessary to wind up its affairs. Bunker and Holmes isolated 1,900 milligrams of radium from the ores and residues which the company had on hand. The Keystone Reduction Company had also developed fine bodies of ore carrying 5 to 6 per cent of uranium oxide, but found it impossible to continue operations at a profit. The Pittsburgh Radium Corporation shipped its ores from Thompson, Utah, to a plant it had established in the old cotton mill in Denver. The W. L. Cummings Chemical Company did not operate during 1923."
>
> In 1924 the United States Radium Co., continued mining and isolated some radium, but in 1925 even this company had to close its plant.

The domestic output has come almost wholly from a relatively small area in southwestern Colorado and southeastern Utah. The principal producing localities are all situated in this irregular area, which extends about 100 miles westward into Utah and 30 miles eastward into Colorado from the State line. Nowhere is it more than about 50 miles wide. A map of this area (fig. 1), published by Barker and Schlundt[44] is reproduced herewith. These authors trace the boundaries as follows:

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[43] Hess, F. L., Rare Metals, Mineral Resources of the United States, 1923, U. S. Geol. Survey, 1927, pt. 1, p. 246.

[44] Barker, Howard H., (with collaboration of Herman Schlundt). Experiments on the Extraction and Recovery of Radium from Typical American Carnotite Ore. Univ. of Missouri, Eng. Exp. Sta. Bull. Series 23, Sept., 1923, p. 25.

> "Limiting it more definitely, the northern boundary starts in Utah and includes Green River, Thompson, and Cisco and continues east and slightly to the south to the Utah-Colorado border, where the Dolores River crosses, and continues up the river to Gateway. From this point the boundary is drawn east to the foot of the Uncompahgre Plateau and thence southeast to include what is known as the Grub Stake Camp, on Mesa Creek. The line then runs a little east of south to the mouth of Tabequatch Creek and thence south into East Paradox Valley, passing east of Thunderbolt Camp. From this camp the boundary runs southwest around the head of Gypsum Valley and Klondyke, and thence southeast to the canyon of the Dolores River, from which point it runs west to the edge of the State and continues toward Monticello, Utah, to include the deposits along the edge of Dry Valley, and thence west toward the junction of the Grand and Green Rivers, and thence southwest to include the Henry Mountains. It is then extended west and north around the San Rafael Swell and joins the points including Green River, Thompson, and Cisco."

From a study of various reports, these same authors summarize the geology substantially as follows: Four formations are exposed in the region namely, Dolores, La Plata, McElmo, and Post McElmo. The Dolores formation (Triassic) consists of 1,000 feet or more of shale and sandstone, with a massive bed near the top. In some places the La Plata formation (Jurassic) consists of one stratum of massive cross-bedded sandstone, but in most places south of the Paradox Valley this formation shows two such strata separated by a few feet of shaly sandstone or limestone. The McElmo formation (Cretaceous or Jurassic ?), which is from 700 to 1,000 feet thick over much of the area, contains at least four, and in most places, five, bodies of very resistant sandstone, separated by layers of shale or limestone. The Post McElmo formation (Cretaceous) is part of the Dakota series of shales and soft sandstones. With but few exceptions, uranium-bearing ores are found only in the McElmo formation and for the most part in two zones,—an upper one, 275 to 325 feet above the base, and a lower one, 60 to 125 feet above the base of the formation. The upper zone, being approximately on the line between the easily eroded shale beds above and the more resistant sandstone beneath, frequently appears near or at the top of the benches that result from the uneven weathering of the formation. The lower productive zone is easily followed because of its association with massive beds of sandstone, separated by lenses and seams of shale and including the first prominent bed of the series above the La Plata formation. At Temple Mountain, however, the uranium and vanadium deposits are in the asphaltite, which is in the Shimarump (upper Triassic) conglomerate,45/ a much older formation, but this occurrence is unusual. The fact that vanadium-bearing minerals are found below the McElmo in the Chinle formation (Triassic),46/ in the La Plata formation at Placerville, and in the Triassic shales of the San Rafael swell indicates that formations lower than the McElmo may contain uranium-radium ores. On the other hand, no uranium or vanadium has been found so far in formations lying above the McElmo series, and prospecting for radium or allied minerals in beds of Cretaceous or later age has been uniformly discouraging.

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45/ Hess, Frank L., Uranium-Bearing Asphaltite Sediments of Utah. Eng. Min. Jour. Press, vol. 114, No. 7, 1922, pp. 272-276.

46/ Gregory, H. E. Geology of the Navajo Country. U. S. Geol. Survey, Prof. Paper 93, 1917, p. 38.

The principal productive deposits in Colorado and Utah are all of carnotite; roscoelite, gypsum, copper carbonates, and rare vanadium minerals may be associated with it. The carnotite occurs chiefly in sandstone. Sometimes it impregnates the sandstone, but it is likely to be concentrated along cracks or bedding planes or in pockets, where it may be accompanied by petrified wood, bones or other more orless fossilized organic matter. Carnotite also occurs in New Mexico and Arizona. The deposits in both these States, though largely undeveloped, may be of considerable importance.

## Other Deposits

Colorado has also produced pitchblende to the extent of perhaps 150 tons or more of low-grade ore, mainly from Gilpin County. Toward the end of 1929 interest in these mines was revived, and one of them was actually being unwatered. Five mines in this area, though worked mainly for gold, have yielded uranium ores. The Kirk, German, and Belshar mines are located on Quartz Hill, and the Ward and the Calhoun mines are in the valley below. The country rock is largely gneiss and crystalline schist intruded by dikes of andesite and acid granite. The ore deposits are of two distinct types; one contains pitchblende associated with pyrite (and sometimes marcasite) and zinc and lead sulphides, while the other contains chalcopyrite along with the other sulphides and some gold and silver, but no pitchblende. In North Carolina pitchblende and certain of its alteration products, such as gummite, have been produced — partly as a by-product of feldspar and mica mining — for many years, but the output has been small and has been purchased almost entirely as mineral speciments and sold at specimen prices. Occurrences in other States appear to be of scientific interest only. Pitchblende is mentioned as a constituent mineral in several quarries in Connecticut (Middletown, Glastonburg, and Branchville); in the Barringer Hill district, Llano Couty, Tex.; and in the Black Hills, South Dakota.

## IMPORTS AND EXPORTS

Before the World War, England, France, and Germany extracted radium from ores imported in large quantities from America, but subsequently all the domestic production was treated in the United States. In 1906 exports of uranium ore, valued at $6,870 (approximately the entire production), were recorded, but data for other years are not available. Investigations by the Bureau of Mines developed the fact that in 1912 carnotite ores, containing 28.8 tons of uranium oxide ($U_3O_8$), and 8 or 9 grams of radium were produced and that almost an equal quantity was sent abroad in 1911. In 1913 and again in 1914, the amount of mineral exported corresponded to almost this quantity of radium, probably 7 or 8 grams each year. Even in 1929 and 1930 inquiries were made by foreign firms for American carnotite, but little or no actual business resulted therefrom. Such ores are low grade; and under existing circumstances if they are to be profitably treated, they must be reduced to less bulky form before they can be shipped vary far. Uranium-bearing residues are probably exported occasionally, but no figures are available as to the quantities so shipped.

In 1920, 396 pounds of uranium ore valued at $1,110 appeared in the import statistics, and in 1921, 250 pounds valued at $446, were noted, but in later years there have been no imports of ore.

Imports of radium salts were almost negligible before 1923, but subsequently they increased to andremained virtually stationary at about 10 grams, valued at a half million dollars yearly. Small amounts of radioactive substitutes (mesothorium) also appear in the import statistics, but only in insignificant amounts The imports of uranium compounds, due to a vogue for brown and yellow glass in recent years and to the absence of domestic production, have increased sunstantially. (7798)

## URANIUM AND VANADIUM ORES SHIPPED FROM MINES IN THE UNITED STATES, 1913 - 1925 1/

| Year | Ores | | Radium isolated grams | Estimated content of ores | | |
|---|---|---|---|---|---|---|
| | Short tons | Approx. value as ore | | Radium grams | Uranium short tons | Vanadium short tons |
| 1913 | 2,269 | $120,000 | - | 10.5 | 34.8 | 432 |
| 1914 | 4,294 | 441,300 | - | 22.3 | 74.0 | 452 |
| 1915 | 47,000 | 693,750 | - | 6.1 | 19.9 | 627 |
| 1916 | 20,300 | 500,000 | - | 10.0 | 33.7 | 460 |
| 1917 | 33,404 | - | 11.96 | 26.4 | 87.4 | 484 |
| 1918 | 18,860 | 957,000 | 21.7 | 27.1 | 89.5 | 276 |
| 1919 | 14,509 | 1,630,900 | 28.648 | 28.96 | 95.7 | 284.2 |
| 1920 | 35,076 | 2,401,000 | 32.539 | 43.4 | 143.5 | 526.7 |
| 1921 | 8,540 | 876,000 | 35.693 | 33.0 | 109.0 | 201.5 |
| 1922 | 929 | 166,016 | 24.189 | 7.0 | 23.2 | 25.9 |
| 1923 | 2,025 | 150,000 | 12.212 | 5.5 | 18.2 | 63.3 |
| 1924 | 2/ | 2/ | 2/ | 2/ | 2/ | 2/ |
| 1925 | 13,070 | 331,000 | - | - | - | - |

1/ Compiled largely by F. L. Hess, U. S. Bureau of Mines.

2/ Bureau of Mines not at liberty to publish figures for 1924 (and 1926 and later years).

## Uranium and Radium Salts (and Substitutes) Imported For Consumption in the United States, 1913-1929

| | Uranium oxide and salts | | Radium salts | | Radioactive substitutes 1/ | | Total |
|---|---|---|---|---|---|---|---|
| | Pounds | Value | Grams 2/ | Value | Grams | Value | Value |
| 1913 | - | 3/$34,319 | - | - | - | - | 34,319 |
| 1914 | - | 3/63,055 | - | - | - | - | 63,055 |
| 1915 | - | 3/17,288 | - | - | - | - | 17,288 |
| 1916 | - | 3/ 5,065 | - | - | - | - | 5,065 |
| 1917 | - | 3/ 2,930 | - | - | - | - | 2,930 |
| 1918 | - | - | - | 274 | - | - | 274 |
| 1919 | 154 | 1,309 | 0.84 | 11,651 | - | - | 12,960 |
| 1920 | - | 3 | - | - | - | - | 3 |
| 1921 | - | 8,552 | - | - | - | - | 8,552 |
| 1922 | - | - | - | - | - | - | 28,685 |
| 1923 | 5,495 | 7,388 | 8.68 | 493,068 | - | - | 500,456 |
| 1924 | 12,550 | 10,161 | 8.10 | 408,146 | - | - | 418,307 |
| 1925 | 19,290 | 18,553 | 9.27 | 462,095 | 0.012 | 241 | 480,889 |
| 1926 | 53,411 | 46,974 | 10.97 | 548,788 | - | 671 | 596,433 |
| 1927 | 67,692 | 58,971 | 7.26 | 393,140 | - | 522 | 452,633 |
| 1928 | 165,378 | 145,557 | 10.97 | 566,977 | - | 2,913 | 715,447 |
| 1929 | 272,913 | 344,548 | 10.69 | 579,085 | - | 183 | 923,816 |

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1/ Classified in 1925 as "bromide of mesothorium and radiothorium."

2/ Reported by the Dept. of Commerce in grains; 1 gram = 15.43 troy grains.

3/ Includes some radium salts.

## THE INDUSTRY IN FOREIGN COUNTRIES

### Australia

South Australia.- Two uranium-bearing areas are known in South Australia, one some 12 miles from Cutana Siding, and 50 miles from Broken Hill, approximately in 140° 40' east longitude and 31° 20' south latitude; the other, which appears to be much more important, is at Mount Painter in the northern Flinders Range, about 90 miles from Copley, on the Great Northern Railway in about 139° 15' east longitude and 30° 20' south latitude. Uranium minerals were discovered at Olary in May, 1906, and at Mount Painter in 1911. Three companies have been formed to exploit the Mount Painter deposits, but efforts at development in both localities have been spasmodic, and due to the difficulty of mining and the cost of treatment the output has been less than half a gram. The proposed merger of the Australian Radium Corporation and the British Syndicate, in 1929, however, was expected to result in a production of about 1 gram of radium a month.

According to a report by Mr. L. J. Winton, Chief Inspector of Mines of South Australia [47] the main workings of the Mount Painter radium field are about 8 miles by road northeast from Copley, on the Great Northern Railway, 374 miles from Adelaide. The radioactive minerals are fergusonite and monazite, and the alteration products are torbernite, autunite, uranophane, gummite, and uranium ocher. Torbernite is most abundant, and autunite next. The field is extensive, but not high-grade. In the past a little hand-picked ore was produced, but the best ore available at the present time appears to contain 1.74 per cent uranium oxide, or approximately 4.7 milligrams of radium element per ton, and most of the ore will show a much lower content. Sampling is difficult, but in the No. 6 workings, where some systematic mining has been done, it is reasonably probable that a few hundred tons of lode material that will average 0.5 per cent uranium oxide ($U_3O_8$) can be extracted. The ore minerals are irregularly distributed. Sometimes they occur in small seams and veins, sometimes as small scales thickly or sparsely disseminated, and sometimes only as a coating along the joint planes of rock. This type of occurrence, of course, renders if difficult accurately to estimate either the tenor or the tonnage of ore that can be mined. Reports from various sources indicate that ore containing 3.3 per cent $U_3O_8$ has been produced, and that it may be possible even from run of mine ore containing 0.5 to 1 per cent uranium oxide to prepare for shipment a concentrate containing up to 3 per cent, or making it equivalent in grade to American carnotite.

Because of the low grade of the ore, the remote location, and heavy transportation costs, it will be necessary to provide for concentrating the ore at the mine, and possibly to produce radium from the concentrates. Research directed toward the latter end has been in progress for some time, and in fact the Dry Creek plant at Adelaide actually produced 104.01 milligrams of radium previous to 1929. The cost is reported at something like £2,500, but according to a director of the Mount Painter Co., improved methods of concentration discovered at the research department of the South Australian School of Mines should make it possible to reduce refining costs by £940 a gram saving, among other things, over 1,500 pounds of hydrochloric acid per ton of ore treated.

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[47] Quoted in Chemical Age, vol. 21, London, Sept. 28, 1929, p. 287.

The radium field at Radium Hill, near Olary, is better situated as regards transportation, and it is close to the Broken Hill Railway. The ore contains titanium, as well as radium, and is considered difficult to treat. It is crushed with sodium carbonate solution in a ball mill and when ground sufficiently fine is discharged and blown to an overhead tank by means of a steam injector. It is digested for hours with boiling sodium carbonate to extract the uranium. The pulp and liquid are then transferred to settling tanks, whence, after a suitable interval the clear solution is decanted, the pulp being again digested with sodium carbonate solution to insure complete extraction. Uranium is afterward obtained as sodium uranate by the addition of caustic soda to the liquor. The residue, after the extraction of the uranium, is well washed to remove alkali and is then boiled with hydrochloric acid to extract the radium, which is precipitated from the solution in the form of sulphate. The sulphate is afterwards converted into a bromide, and the radium bromide is finally obtained in the required degree of purity by repeated fractional crystallization in the usual way.

West Australia.- Mackintoshite, thorogummite, and pilbarite occur in a pegmatite dike at Woodgina, and occurrences of fergusonite and euxenite have been reported at Cooglegong. According to the British Radium Subcommittee, it is unlikely that any considerable production can be expected from this source.49/

## Belgian Congo

The relative abundance and richness of the Congo ores have given the Belgians a practical monoply of the radium industry since 1924; and although it is one of the more recently exploited fields, the Belgian Congo has already furnished substantially one-half of the radium that has been offered to the world in usable form.

Uranium minerals were discovered in Katanga in the course of prospecting for copper. The first discovery was made on January 22, 1913, at the Luiswishi mine, and the second on April 10, 1915, at the Chinkolobwe (Kasolo) deposit. 49/ The latter deposit, which is about 125 kilometers as the crow flies from Elizabethville, is the only one that has been exploited for radium ores. In common with certain other products (including tin, cobalt, and gold) of the Union Miniere de Haut Katanga, this ore is refined in a plant of a sister company, the Société Generale Metallurgique de Hoboken, at Oolen, near Antwerp. The first lot of Katanga radium ore, about 12 metric tons, arrived at Antwerp on December 5, 1921. Construction of the radium plant was started in March of the following year, and a section was placed in operation on July 8. The first tube of radium from this new source appeared on the market in August, 1922, and by the end of the year about 6 grams of the element had been produced. The first 8 grams were placed at the disposition of the four Belgian universities to be employed for scientific research and therapeutic uses. Production reached the rate of 2 or 3 grams a month for a time but dropped off as stocks began to accumulate. The plant was closed in 1926 and did not reopen until October, 1927. In that year, however, sales jumped to 28 grams, and in 1928 to 40 grams; in 1929 both sales and output are believed to have further increased, greatly exceeding all previous records. Several years ago the plant was reported to have a capacity of 50 to 60 grams a year, which could quickly be enlarged should market conditions demand it.

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48/ Committee of Civil Research, Radium Subcommittee Report (Cmd. 3303) London, Mar., 1929, p. 15. 49/ Sengier, E., La decourverte dy minerai de radium au Katanga. Communications, etc.,at Colonial Exhibit at Tervueren, Nov.13, 1922, pp. 23-29.

The ore at Chinkolobwe is pitchblende accompanied by chalcolite (torbernite), curite, and kasolite. The last two are more or less peculiar to the district; both are uranates of lead, and the last also contains silica, which replaces part of the uranium oxide. In the thickest portions of the veins pitchblende appears at the center, and the torbernite and other altered minerals are near the walls. The veins are narrow and very irregular in strike, dip, and thickness, alternately pinching and swelling, and they branch and disappear into mere cracks. All mining is open-cut, and little or no development is carried on in advance of mining. In view of the nature of the occurrence, therefore, no reliable estimate can be made as to reserves. The country rocks are sedimentaries, largely dolomitic limestones and shales, and they are highly metamorphosed. Many of the vein outcrops carry cobalt and copper.

The ore is loaded at the mine into sacks, which after being hauled 20 kilometers by motor truck are placed in cases and shipped by rail and steamer, passing through Rhodesia to the Port of Beira on the east coast. Four white men and about 200 natives constituted the working force in 1922.

The uranium oxide content of the ore has been as high as 60 per cent, and originally the ore was so rich that only 10 tons had to be treated in order to produce 1 gram of radium. In 1928, however, about 30 or 40 tons of hand-picked ore, as shipped, was necessary to produce 1 gram.[50]

## Canada

Radium-bearing pegmatites have been noted from time to time in the various district in eastern Canada. None of these earlier discoveries, however, seemed to have commercial possibilities although serious consideration was given to an occurrence of euxenite which was found in some quantity at a feldspar quarry in South Sherbrooke Township, Lanark County, Ontario, in 1917. A few hundred pounds of concentrates were made at the Government laboratories from a small shipment of this ore in 1919 but since euxenite is rather low in uranium (7.29 per cent $U_3O_8$) no attempts were made at commercial production. Suggestions have been made that certain low-grade ore containing only a pound or two of uranium oxide to the ton might under certain conditions prove workable, more particularly when the radium minerals could be produced as a by-product of mica or feldspar mining.

Recently, however, prospecting in the Wilberforce district of central Ontario has resulted in the discovery of a pegmatite dike carrying uraninite which appears to have considerable promise. This dike, which outcrops about 1 mile east of the village of Wilberforce in Haliburton County, has been traced for several hundred feet. The ore is disseminated in the form of large crystals and lumps in a lead of fluorspar which also carries large crystals of apatite. This lead is believed to have a length of at least 500 feet and runs in width from 5 to 12 feet.[51] Tests by the Canadian Government laboratories showed that 1 gram of radium would be recoverable from the concentrates obtained from 3,422 tons of ore. The $U_3O_8$ content of the concentrates obtained from 1 ton of ore was 2.56 pounds.

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[50] Committee of Civil Research, Radium Subcommittee Report (Cmd. 3303). London Mar., 1929, p. 17. [51] Spense, Hugh S., and Carnohan, R. K., The Wilberforce Radium Occurrence. Canadian Mining and Metallurgical Bull. No. 217. May, 1930, pp.649-688.

## Czechoslovakia

The first radium produced by Madame Curie was from pitchblende mined at Joachimsthal; and previous to the rapid development of American carnotite deposits, about 1914, the bulk of the world's supply of radium came from ores mined in Bohemia. The mining town of Joachimsthal (Joachim's Valley) was named for Count Joachim Schlik, who exploited the silver mines so successfully that the coins minted from th metal produced in that locality became famous for their genuineness and standard value. They were first called "Joachimsthalers," but this was soon abbreviated to "thaler," perpetuated in the name "American dollar." Joachimsthal, or Jachymov, as it is called by the Czechs, is situated 16 kilometers north of Carlsbad in Ore Mountain (German, Erzgebirge; Czech, Krusny Hory) a moderately low range of mountain that separates Bohemia and Saxony. It was here that Agricola spent six years (1527 to 1533) and secured much of the information for his pioneer treatise on mining, De Re Metallica, which was translated from medieval Latin some years ago by Herbert and Lou Henry Hoover.

These famous mines have been worked since 1512. In 1545 the silver output declined considerably, and later the silver resources became virtually exhausted. Mining continued in a desultory fashion and was revived on a small scale as a source of bismuth and cobalt for ceramic colors. Uranium was found in the middle of the last century, and a few of the old mines were reopened to supply this material for c coloring porcelain; but during the last 25 years radium has been the principal sourc of revenue. The Austrian Government built a factory at Joachimsthal to treat pitchblende and in 1912 obtained control of the mines. These enterprises became the property of the Czech Government after the World War, and operations have been continue by an Anglo-Czechoslovakian company, under a working agreement with the Government. Some progress has been made in modernizing equipment. Before the War a substantial tonnage of American carnotite was treated by the Joachimsthal plant.

The deposits are in mica schist, interbedded with limestone. To the east and northeast is a formation of gneiss, which was intruded by quartz subsequent to the deposition of vein material. The fissures in the mica schist are filled with volcanic material, which cuts the mineralized zone at various points and depths. Th mineralixation is confined largely to a zone about 6 kilometers square and is probably of Cretaceous or post-Cretaceous age. There are two series of intersecting veins. One series (Morgen viens), trending east and west, contains no uranium and pinc es out at depth; the other series, trending north and south, is uranium-bearing and continues to considerable depths-- in fact, the uranium in these veins tends to increase with depth. Geistergang vein, for example, has been worked from the time tha mining began in the district and the Werner mine uranium was encountered in only sma amounts (associated with cobalt, lead, and bismuth) in the upper levels, but below 262 meters it appeared in increasing quantities, and below 376 meters it became the only valuavle constituent of the ore. The veins range in width from 6 to 20 inches, rarely reaching 3 feet; the ore occurs both in stringers and in pockets. Three peri of deposition are indicated. During the first period cobalt and nickel minerals wer deposited, next uranium, and lastly silver. Dolomite is always present; it is generally yellowish-white in color, changing to brownish-red in the vicinity of pitchblende, and is dirty gray in actual contact with the ore. Deep-blue fluorspar is also present. In places the pitchblende is not confined to the vein but is disseminat as small grains in mica schist in sufficient concentrations to form low-grade ore.

Since the veins occur along the ridge, some 400 meters above the valley, they are favorably situated for mining.

Production from 1920 to 1924 has been as follows:

| Year | Grams |
|---|---|
| 1920 | 2,257 |
| 1921 | 3.221 |
| 1922 | 2,256 |
| 1923 | 1.521 |
| 1924 | 1.433 |

During 1925, mining of radium ores at Jachymov (Saint Joachimsthal) continued in spite of the difficult market. The Werner, Svornost, and Shaft, of the Saxon Nobility mines, were worked at depths of 120 to 150 meters, and 1.2 grams of radium were isolated. During 1926 the radium and uranium works were shut down to allow enlargement and improvement of the plant. The mines produced 103 tons of uranium ore, giving 22 metric tons of pitchblende, carrying 50 per cent uranium oxide ($U_3O_8$), and thus presumably containing about 2.82 grams of radium. It was expected that the radium production for 1927 would be about 3 grams, although when completed the plant would have a capacity of 6 grams a year.

The Government owns and operates the mines and has heretofore held the radium for research, educational, and clinical uses. Sales to private individuals through the Government-owned mining and metallurgical company were begun in October, 1927, but according to a British Government report,[52] published in 1929, exportation of radium from Czechoslovakia is still forbidden. A few years ago 1.6 grams were sent to an English company, which was to use it for therapeutic purposes. The company failed, and the radium was deposited with the Czechoslovakian legation in London. The Government has sold some radium, but the remainder has been rented to physicians at 10 per cent interest or deposited in the Government's banking office at the same rate.

The output of uranium compounds for ceramic colors is estimated at 13,000 kilograms (28,600) pounds annually, at prices ranging from $2.75 to $3 a kilogram.[53] There are 30 plants in Jachymov for making pigments, mostly for glass and pottery coloring.

During the latter part of 1924, the Mining Journal of London [54] said:

> "It is reported that, owing to the diminished demand and to the discovery of radioactive ore in America and the Congo, a loss of 80,000,000 kronen has been made on the Joachimsthal stock of radium, the price of which has fallen from 11,000,000 to 3,000,000 kronen per gram, and that the Czechoslovakian Government have abandoned the project of a State monoply."

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[52] Committee of Civil Research, Radium Subcommittee Report (Cmd. 3303). London, Mar., 1929, p. 18.

[53] Hess, Frank L., Rare Metals. Mineral Resources of the United States, 1927, pt. 1, Bureau of Mines, 1929, p. 443.

[54] Min. Jour., vol. 157, London, Oct. 25, 1924, p. 839.

During 1923 local Czech papers reported that deposits of great promise were discovered at Durrmaul, in the region west of Marienbad, on the east slopes of the Bayerischer Wald. 55/ Pitchblende was found at a shallow depth in certain old copper-bearing properties that have long lain idle. The ore is stated to average 5.5 per cent of uranium, or 0.0177 gram of radium to the ton. A joint stock company, the Bohmische Montan-Aktiengesellschaft, has been organized with a stock capital of 11,000,000 Czech crowns, for the purpose of developing the deposits; the headquarters of the company are at Hotel Neptun, Marienbad. Before the end of 1923 three properties in this region had been surveyed and granted by the Government for purposes of development.

## France

Although no radium ores have been produced in France, the country is more or less self-sustaining as regards its supply of the element. Radium-extraction plants have operated fairly steadily for many years. Armet de l'Isle, at Nogent-sur-Marne, and the Henri de Rothschild, on Isle de Saint Denis, have been producing radium from imported ores, chiefly betafite from Madagascar and pitchblende from Portugal. For a time (1910-1911) torbernite was obtained from French Indo-China, and most of the carnotite exported from the United States before the War was treated in France. The plant at Nogent was built in 1904, but the one at Saint Denis was built subsequently, especially to treat Madagascan ore.

A discovery 56/ of radium minerals at Saint-Remy-sur-Duralle was announced in 1927, but nothing further seems to have been heard of it.

Discoveries of torbernite (chalcolite) deposits have been reported from the Puy de Dôme region of France,57/ and a company was reported to have applied for a permit to work lead, zinc, copper, and radioactive ores in the Villefranche Arrondissement Rhone Department.58/

## Great Britain

Radium has been produced at various times in several different mines in Cornwall. For many years the pitchblende, torbernite, autunite, and zippeite, associated with copper ore, were regarded as undesirable gangue minerals, but later they were recovered in somequantity both from underground and from the dumps. The mines appear to have been exploited for radioactive substances with some regularity. The principal one is the South Terras mine, situated in the valley of the River Fal, 5 miles due west of Saint Austel and 2 miles by road from Grampound Road station. The other is the Saint Ives mine or Trenwith mine), near the seacoast, 2 miles from Saint Ives, which has apparently been abandoned. It was operated by Saint Ives Consolidated Co., radium being made by a subsidiary company, the British Radium Corporation, at Limehouse, London. The total output of uranium or radium from Cornwall is not known, but a British authority 59/ asserts that at least 637.5 long tons of uranium ores was produced and sold from the Saint Austel area. Of this amount 440 tons was sold ofr L41 a ton; 195 tons picked off the dumps brought an average of perhaps L37 a ton, and for 2½ tons no price was published. An additional quantity was picked

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55/ Yoder, W.A., New Radium Deposits Discovered in Czechoslovakia. Dept. of Commerce Trade Note 91, Feb. 5, 1924.

56/ L'echo des mines et de la metallurgie, Decouverte de minerai de radium. Vol. 55, Aug. 1, 1927, p. 343. 57/ Min. Jour., vol. 163, London, 1928, p. 952. 58/ Min. Jour., vol. 162, London, 1928, p. 806. 59/ Worsley, A., Report of the Radium Subcommittee (Discussion). Min. Jour., vol. 165, London, April 27, 1929, pp. 329-330.

from another mine in the same district, but there is no record of the tonnage. The maximum price for the South Terras ores was £100 and the minimum £20 a ton.

The radium lode (strike north) at South Terras mine was first encountered in 1873 while working on an iron lode (strike northwest) near its intersection with a tin lode (strike northeast). As the uranium content was sufficient to warrant exploitation, stoping was begun in 1878 above the adit level, and later one and then another shaft was sunk. Shortly after 1900 the mine was closed, as all the ore above the 20-fathom (120-foot) level had been worked out. It was reopened in 1906, but deeper exploration indicated that the known ore shoot became exhausted at about the 30-fathom level. In 1909 this shoot was worked out, and development (always costly because of high pumping costs) was stopped. The ore was exported for use in the manufacture of glass. Two grades were sold, one averaging about 20 per cent and the 11 per cent uranium oxide ($U_3O_8$). These grades were made up by mixing high-grade ores containing 36 per cent uranium oxide, with low-grade ores containing as low as 3 per cent; the latter figure appears to represent the minimum workable limit for the ore.

At the beginning of 1913 the property was taken over by the Société Industrielle du Radium (Ltd.) to recover the radium content, and it was then estimated that the dumps and the stope fillings contained in all some 36 grams of radium element. During 1913 and 1914 the dumps were picked over, and the best material was sent to France for treatment. No work was done after 1914 until 1920 and 1921, when £3000 to £4000 worth of mineral was sold from the stope filling above the 10-fathom (60-foot) level and shipped to France. The remaining ore being considered too low-grade to ship, a treatment plant was erected at the mine in 1922, and radium was produced from March, 1923, until the end of 1925. In 1927 the present company, the British and General Radium Corporation (Ltd.), took over the property with the object of prospecting it at depth. All the known payable uranium ore from this lode was obtained from the one ore shoot. Exploratory work elsewhere in the mine has shown sporadic values of radium, but the average run has been too low-grade to work. The future of the mine, therefore, depends on the discovery of additional shoots.[60] Prospecting was in progress in 1928 and 1929.

A British Government report[61] says, "We think it improbable that any of the Cornish ores are sufficiently abundant to enable radium to be extracted from them in commercial quantities in competition with the ores of the Belgian Congo."

According to Hess [62] the Cornish production from 1913 to 1922 was as follows:

Uranium ore produced in Cornwall, 1913-1922, in metric tons [63]

| | | | |
|---|---|---|---|
| 1913 | 97 | 1917 | 13 |
| 1914 | 350 | 1920 | 61 |
| 1915 | 83 | 1921 | 77 |
| 1916 | 52 | 1922 | 399 |

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[60] Robertson, T., and Dines, H.G., The South Terras Radium Deposit , Cornwall. Min. Mag. vol. 61, London, Sept., 1929, pp. 147-153.

[61] Committee of Civil Research, Radium Subcommittee Rept. (Cmd.3303). London, Mar., 1929, p. 14.

[62] Hess, Frank L., Rare Metals. Mineral Resources of the United States, 1926, U. S. Geol. Survey, 1927, pt. 1, p. 254.

[63] None produced in 1918 or 1919.

## Madagascar

The production and exportation of uranium ores in Madagascar began in 1912, in which year a shipment of 11 kilos was made. In 1922 exports had increased to almost 16 metric tons, and in the following year 15 tons of betafite and 10 tons of euxenite and monazite were reported as exported. After dropping to about 5 tons in both 1924 and 1925, and to 2 tons in 1926, the exports dwindled to only 158 kilograms in 1927, and ceased entirely in 1928. Madagascan ore has all been shipped to France. Exports to countries other than France or French possessions were prohibited by French presidential decreee dated August 6, 1922.

Radioactive minerals are said to be found in Madagascar that carry more than the normal quantity of radium.[64]

The radium-ore district (Sama), in the vicinity of Lake Itasi, is about 200 kilometers west of Tananarive, which in turn is 350 kilometers from the coast. Transportation to Tananarive is accomplished on men's backs at a cost of 100 francs per ton. Freight by rail from Tananarive to Tamatave amounts to 150 francs per ton, and ocean freight from this port to France is approximately 100 frans per ton.[65]

## Portugal

Radium ores have been mined in Portugal for a number of years. The Portugese ore is low-grade, and the radium content is substantially less than the normal equilibrium ratio with regard to the uranium content, but it has the reputation of being relatively easy to treat. A little of this ore has gone to England, but the bulk of it has been treated in France.

Portugese production data are fragmentary and generally relate to ore of which the assay is not stated. In 1919, 1,380 metric tons of ore was produced; in 1920, 437 metric tons; in 1921, 1,032 metric tons; in 1922, 945 tons; in 1923, 355 tons; and in 1924, 367 metric tons. In 1925, when 240 tons was produced and concentrated, "part of the concentrates were treated in Portugal, where 0.875 grams of radium was isolated and the remainder, 180 kilograms containing 0.360 grams of radium was shipped to France. A stock of about 1,600 tons of vein material, carrying 0.5 to 0.7 per cent uranium oxide ($U_3O_8$), is believed to be available.[66] No ore was produced in 1926. In 1927 only 0.2 metric tons were reported, but in 1928 the output increased to 144 tons. In 1929, at least two concessions were being held by British companies, and there was some discussion as to the prospects of renewed mining activity.[67]

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[64] Lacroix, A., Les industries minerales nonmetalliferes à Madagascar. Bull. écon. de Madagascar, vol. 17, 1920, pp. 48 et. seq.

[65] These figures, which are given by Maurice Curie (Le Radium et les Radio-éléments, Paris, 1925, p.160* apparently relate to 1922, for which year the average exchange value of the franc was 8.2013 cents.

[66] Committee of Civil Research, Radium Subcommittee Rept. (Cmd. 3303). London, Mar., 1929, P. 18. (It should be noted that the radium produced in 1925 was probably in excess of that which would be yielded by the ore produced in the same year.)

[67] Mining Journal, Compare Radium and Uranium Ores in Portugal. Vol. 165, No.4886, London, April 13, 1929, p. 279.

According to Segaud and Humery, 68/ uranium veins are persistent but irregular in width and extent of mineralization. One vein 8 meters wide has beem t traced for a short distance, but a width of $\frac{1}{2}$ to 1 meter is more common. The bedrock in the region consists entirely of massive granite, which occupies the entire northern part of Portugal between the Desert of Galice and Castello Branco, and which reaches into the Provinces of Minho, Tras-os-Montes, and Beira. The richest portion lies between the towns of Guarda and Sabugal, near the southern part of the granite area, in places touching the outcrops of Cambrian rocks (north of Guarda), and also in the zone south of Guarda, where the granite forms a superficial mass which has been thrust above the sedimentary rocks by faulting. The whole region is highly mineralized and shows deposits of galena, arsenopyrite, and cassiterite. Wolframite occurs to the north of Guarda. The region of Guarda is an elevated plateau, having an altitude of 2,700 feet, dotted with small villages and traversed by a railway from Pampilhosa to Villar-Formosa and from Guarda to Lisbon. In general the ore is lean. Material containing 0.3 per cent to 0.5 per cent has been considered profitable to mines, and 1 per cent ore is classed as good and 2 per cent as excellent. Efforts made to concentrate the ore chemically at the mines proved successful for black minerals but not with typical autunite-torbernite material.

Some of the ore or concentrates shipped in recent years to France is said to have carried as much as 16 per cent uranium oxido, but most of that which has been shipped seems to have carried about 1 per cent. In 1913, the principal workings were those of the Compagnie de l'Urane, which employed 600 men. Small drifts, $3\frac{1}{2}$ by $4\frac{1}{2}$ feet, were driven for about $17 a yard, and the average net cost of broken ore was reported by Segaud and Humery to be about $20 a cubic meter, which is probably equivalent to $10 a ton. At that time the ore was treated chemically at the mine without previous mechanical concentration, yielding two products, uranium oxide and radium-bearing sulphate of barium. The latter was shipped for further treatment in factories at Nogent.

The following notes on the geology of the region were digested by Hess, 69/ largely from an article by Duparc.

> "The granite is of two varieties; one contains 67 per cent of silica, carries black mica, and is somewhat porphyritic; the other is a granulite carrying 70 to 74 per cent of silica, with muscovite or with two micas, and the quartz has a bipyramidal tendency. Quartz veins that carry wolframite and cassiterite and those that carry radium minerals occur only in the second type.
>
> "The veins form a series of parallel groups, which successively eastward from the Sierra Caramullo (Duparc says from east to west, evidently an error), are as follows:
>
> "Sega group, immediately east of the Sierra. Includes the veins of Menninho and Morengueiro, on which little important work has been done.
>
> "Sabougosa group, of numerous known veins but little exploitation.

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68/ Seguad and Humery, Deposits of Uranium in Portugal. Ann. des Mines, ser. 11, vol. 3, Feb., 1913, pp. 111-119.

69/ Hess, F. L., Rare Metals. Mineral Resources of the United States, 1923, U. S. Bureau of Mines, pt. 1, 1927, p. 253.

"Cannas-Senhorim-Cabanas group, on which is the Urgeirica mine and other smaller workings.

"Mangualde group, farther east.

"Maceira-Troncoso group. One vein is worked.

"Guarda group. This is the most eastern group.

"In the outcrops autunite and torbernite are found in the inclosing granite but not in the veins, and the exploitable parts of the veins are indicated by these minerals in the granite. The granite is decayed and porous where impregnated with uranium minerals. The fissures in which the veins are found are attributed to contraction of the granite and in less degree to "dynamic phenomena." In places gouge accompanies the veins. The depths to which the deposits are unknown, but the Urgeirica mine is said to be worked down to 100 meters. Autunite and torbernite are found for 15 to 20 meters from the surface, below which is a barren zone of a few meters; under the barren zone a black or grayish amosphous mineral is found persistently, which is richer in depth."

## Russia

The exclusive right of development of radioactive raw materials in Russia has been granted to a rare elements cartel, which is called the Rjedkij Elmenty, and which has been assigned all rights to prepare, sell, or work with such raw materials. The source of Russian radium is the Tyuya-Muyun deposit situated in the foothills of the Altai Mountains, in the southeastern part of Fergana, in Russian Turkestan, approximately 60 kilometers east of Skobelev, 30 kilometers southwest of Osha, and 55 kilometers south of Andijan.70/

The deposit consists of four barite veins carrying uranium, vanadium and copper combinations, three copper-barite veins, and more than 30 barite veins containing no metallic-ore minerals. These veins form a system that has been traced along the ridge for a distance of about 3 kilometers. The ore-bearing veins follow the center of the field along a strike bearing north 70° east. The ore is very complex. Calcite constitutes about 80 per cent of the vein matter, and clays and ore minerals only 4 per cent. The latter consist mainly of uranovanadates and carbonates, together with a certain amount of sulphides and silicates. The uranovanadates are represented in part by tyuyamunite and in part by an amorphous brown ore of variable composition, which contains an abnormally high percentage of radium in proportion to its uranium content. The vanadates, which are secondary minerals, are represented chiefly by tangeite (or "tangueite") and its earthy varieties (called tobacco ore). Soviet geologists ascribe the deposit to the action of ascending vadose waters, which dissolved ore minerals from underlying shales and redeposited them higher up, chiefly along the walls of caves and crevices.71/

The Fergana deposit was discovered many years ago and was worked by the Chinese in a primitive way for copper. About 1904 it was taken over by the "Fergana Co. for the Production of Rare Metals," which previous to 1914, when it ceased operations, produced about 1,000 tons of ore. Of this amount 700 tons were utilized

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70/ The geographic position of Tyuya-Muyun is 40° 21' north latitude, 72° 35' west longitude from Greenwich (42° 15' east from Pulkovo).

71/ Kirikov, A.P., The Tyuya-Muyun Radium Deposit. Russia Geol. Com. Mem., New Series, No. 181, Leningrad, 1929, 65 pp. (In Russian; English summary, pp. 63-65).

for the extraction of copper, uranium, and vanadium; and the remainder (including certain residues), having an aggregate content of approximately 2.3 grams of metallic radium, was worked up in the Government radium plant erected at Kane under the supervision of the Russian Academy of Science.

An expedition sent to devise ways and means for resuming work at the mine reported in 1922 at least 5,000 tons of developed ore, of which about 1,600 tons were in the upper levels, about 800 tons in a large cave that crosses the shaft, and about 2,600 tons in the lower level. These reserves represent from 15 to 20 grams of radium element, about 60 tons of uranium metal, 90 tons of vanadium metal, and 120 tons of copper. These figures represent only the reserves indicated as "in sight" and hence do not at all represent the maximum yield that might be obtained from the mine.

Other occurrences in Russia are as yet of no commercial consequence. Westward from the Fergana deposits, S. M. Kurbatov [72] found uranium-vanadium minerals in small quantity on the right bank of Bolshaya Erba River, 2 miles south of the village of Potekhino, in the Minusinsk district of the Yenisei Government. Doctor Fersman, [73] A. Lubunzov, [74] and their parties searched for radioactive minerals in Carelia (on the Arctic coast, east of Sweden) and found uraninite in several pegmatites. The total is probably small and scattered as in the pegmatites of Canada or the United States..

## South Africa

There have been several rumors of radium finds in south and southwest Africa, one of which was a discovery of pitchblende in 1926 near Messina, in the extreme north of the Transvaal; and early in 1929 news came of a definite discovery believed to be of economic importance. A syndicate of Johannesburg mining financiers investigated the Gordonia pegmatite areas in Bechuanaland, from which pitchblende samples had been taken, but after doing a little work apparently gave up the project. A later plan was to combine the exploration of various areas in southwestern Africa, and it was stated that a large company was being financed for that purpose, but the situation seems to be summed up in an editorial in a local trade journal [75] to the effect that "it is questionable whether sums running into six figures can be obtained in the ordinary way for exploiting South Africa radioactive ores."

Dr. S. H. Haughton, of the South African Geological Survey, describes the Gordonia radioactive deposits in Cape Province as existing in the form of pegmatite dikes that cut the granite underlying the upper sedimentary rocks and surface sands. The deposits are very limited in number, although the pegmatite dikes are numerous. The discoveries are on farms within 35 miles of the railroad. Samples produced were high in radium content, though scarcely as rich as the Congo ore.[76]

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[72] Kurbatov, S.M., New Deposits of Combined Uranium and Vanadium in the Minusinsk District of the Yenisei Government. Acad. Sci. Bull. 19, Leningrad, 1925, pp. 315-322. (In Russian.)

[73] Fersman, A.E., and Labunzov, A., Report of the Expedition to the Uranium Deposits in Russian Carelia. Compt. rend. Acad. Sci. Russia, 1925. (In Russian.)

[74] Labunzov, A., Deposits of Uranium Minerals in the Carelian Republic. Compt. rend. Acad. Sci. Russia, 1925. (In Russian.)

[75] South African Min. Eng. Jour., vol. 40, July 13, 1929, p. 565.

[76] Letcher, Owen, Johannesburg Letter. Eng. Min. Jour., vol. 127, Mar. 9,1929, p.41

## PRICES

During the World War the price of radium reached $125,000, and occasionally $135,000 or more a gram. Even now the price is $70,000 a gram, except in case of large orders for chartiable institutions. Radium, therefore, is one of the most precious articles of commerce, costing many times as much as ordinarily fine-quality gems. Emeralds and rubies rarely are worth more than $1,000 a carat, or $5,000 a gram, and diamonds as a rule are worth even less. Gold is worth only $0.6645 a gram.

The following table prepared by Professor Camille Matignon 77/ gives the world prices of elemental radium in United States dollars per milligram (one thousandth of a gram) practically from the time it first became commercially available:

World prices of elmental radium per milligram, 1904 - 1930

| | | | |
|---|---|---|---|
| 1904 | $10 - $25 | 1911 - 1912 | $150 |
| 1905 | 25 - 50 | 1912 - 1914 | 180 |
| 1906 | 60 | 1915 - | 160 |
| 1909 - 1910 | 75 - 135 | 1916 - 1922 | 120,110,105 |

1923 (to date) $70

The price of radium is quoted in terms of the actual weight of the element present in a purified radium salt. Radium preparations of all kinds are ordinarily sold on the basis of Government certification as to radium content as determined by gamma-ray measurements. In the United States this work is carried out by the Bureau of Standards of the Department of Commerce, at Washington, D. C. Preparations purchased by doctors or hospitals may be resold or returned to the laboratories for conversion into some other form of preparation, or they may be changed from one type of container to another. Such transactions likewise are ordinarily routed through the Bureau of Standards. Radium must always be handled in sealed tubes, and a great variety of different containers or applicators are demanded, almost every doctor having his own ideas as to the best form. Under these circumstances it is obvious that the cost of handling individual sales is extremely high. It is only reasonable that lower prices should be charged for sales of 1 gram or more in one lot, particularly when such sales have a large advertising value. For the 1 gram donated to Madame Curie by American subscription in 1929, $50,000 was paid, although the nominal price charged for regular business in the United States was still $70,000, as indicated in the foregoing table. Sales in Europe have been reported at $56,000 a gram, and in connection with the proposed program of large purchases by the British Government a price under £1,000 was discussed.

Radon, or radium emanation, is usually sold in the form of gold implants or "seeds," 5 to 7 millimeters long and 0.75 in diameter. Platinum implants also can be obtained. The average price of the gold implants in the vicinity of New York is about $4, per seed of 1 millicurie activity. Tubes, having an initial activity of about 50 millicuries may be rented for about $25 per twenty-four hours.

The prices of radium ores will be discussed under the head of cost of production.

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77/ Work cited.

## COMPETITIVE SITUATION

### General Statement

Since 1923 the world supply of radium has been furnished almost exclusively by a Belgian company, a subsidiary of the Government-controlled Union Miniere de Haut Ktanga, known as Radium Belge. When this company cut the price of radium in 1923 from $100,000 (or more) to $70,000 per gram, it was freely stated that the latter figure was approximately the cost of producing and selling radium derived from available American carnotite ores, while the cost of production from high-grade Congo ores was reported to be not more than $15,000 or $20,000 a gram. The larger American companies accepted this situation, and in order to salvage their elaborate marketing organizations entered into agreements to act as selling agents for the Belgian product. The United States Radium Co. continued to extract radium for a few years in a small way, but all other American firms ceased production as soon as their accumulated stocks of ore were used up. Subsequently, except for 2 or 3 grams furnished annually from the Czechoslovakian Government mines, radium has constituted virtually a Belgian monopoly. After a lapse of some seven years there came a revival of interest in radium deposits elsewhere in the world. The opinion has gained ground in some quarters that the deposits controlled by the Belgians are not rich and extensive enough either to satisfy indefinitely the world's needs or to preclude a chance of radium being extracted profitably from sources outside of the Congo. A British Government subcommittee has officially expressed the opinion that it may prove possible to produce radium from other fields in commercial quantities at an economic price,[78] but adds: "From the evidence before us we are, however, satisfied that sufficient data are not at present available to enable any definite prediction to be made on this point." About the middle of 1929, and only a few months after this report was made public, the English and Overseas Radium Merger (Ltd.) was incorporated in London, with an authorized capital of £1,500,000, to be raised by sales of stock. This new concern, it appears from newspaper reports, is preliminary to a merger of groups controlling radium-bearing properties in Cornwall, South Australia, and Czechoslovakia. The syndicate behind this proposed consolidation has acquired (or controls under option) (1) from the Australian Radium Corporation the mines and mining properties and extraction works in the Mount Painter districts in Australia; (2) the mining leases, mines, and plant of the Radium Ore Mines (Ltd.) situated on Crow Hill, Cornwall; and (3) various properties at Weipert, Czechoslovakia, from the Argenta Silver Mining Company, (a cooperative miners' union) adjacent to the well-known radium mines at Jachimov. Negotiations have been undertaken for the exclusive selling rights of the Czechoslovakian Government mines. In addition to producing and marketing radium, the plans include the construction and operation of a radium "hydro", or spa, in the Fal Valley near St. Austel, utilizing the curative waters from a radioactive spring.

Other radium projects are probably likewise under way, as for example, the test work on the newly dsicovered Canadian ores. In the United States there is renewed activity in the carnotite fields, but at the time this paper is being written (April, 1930) no definite conclusions can be drawn as the prospects of reviving a domestic radium industry.

Although certain fundamental data are lacking, it may be profitable nevertheless to review briefly the various excellent estimates of probable cost.

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[78] Radium Subcommittee Report (Cmd.3303). London, March, 1929, p. 21.

Before discussing costs, however, it may be well to mention briefly certain other important competitive factors in the light of the information that is available. In the first place, the demand for radium can not be predicted with accuracy. Sales of radium have increased quite rapidly in recent years, but is is not possible to hazard even an intelligent guess as to how much radium could be sold in any future year or at what price. To the writer it seems doubtful if a reduction in prices would greatly increase sales; temporarily it might even induce prospective purchasers to postpone buying in the hope of further reduction in prices. Even a charitable institution can scarcely be expected to invest in radium unless there is a reasonable assurance that its stock of this highly costly element will not depreciate too rapidly in value. For the purpose of discussion one might assume that during the next few years the world demand will amount to 40 grams or 50 grams, or that some other fairly substantial quantity of radium might be sold at a price of say $50,000 a gram. Even then, however, it does not follow that the Belgian company would not choose to cut the price down to, or even below, its actual cost of production (another unknown factor) rather than lose business to a competitor.

Domestic ore reserves are not definitely known, but there is reason to believe that a fairly large supply is available, and this is certainly true of low-grade ore. Quantitative data as to the extent and grade of certain deposits are in the hands of private companies, but there are no dependable figures for reserves in any very extensive area. The erratic nature of occurrences and the fact that most of the deposits are difficulty accessible tend to make it almost impossible to prepare a reliable estimate without an extraordinary amount of active field work by competent engineers.

## Cost of Ore

The sale price of uranium-radium ore is variable, and depends upon a number of factors. Formerly sales were based upon the content of $U_3O_8$ and for convenience this method is still in use in connection with the small sales of carnotite in the United States. One kilogram of uranium element corresponds ordinarily to 0.34 milligrams of radium element, but as noted elsewhere in this paper this ratio is not always constant. In the case of autunite, for example 1 kilogram of uranium represents only 0.25 milligrams of radium. Sales, therefore, are more often based on the number of milligrams of radium actually present. The price paid per milligram is not the same for all types of ore, but is scaled according to the richness of the material, ease of treatment, thorium content (which ought always to be relatively small as compared to the uranium content), and of course, market fluctuations. According to Curie (p. 169), the sales price per milligram of radium element contained in ore ranges between one-eighth and one-quarter the price quoted for radium in the form of its salts; it may be greater in the case of exceptional ores or chemical concentrates (radium-barium sulphates).

Vanadium is present in carnotite ores, but even when recovered it rarely is paid for. This seems due partly to the comparatively high cost of recovering the vanadium, and partly to the fact that high vanadium carnotites are more difficult to treat for radium, Mesothorium is extracted almost wholly from the sulphate residues after extracting thorium from monazite. Since the mesothorium is recovered only in the same plants which produce the residues there is scarcely any sale for thorium-bearing ores or residues. Uranium ore containing substantial amounts of thorium - for example, certain euxenites - are actually not marketable.

Most of the radium-producing companies have obtained their ore from their own mining operations, and for this reason relatively little ore has come upon the open market. In 1929 and 1930 small amounts of carnotite ore containing up to 8 per cent of uranium oxide were sold to makers of radioactive drinking water containers for therapeutic purposes for around $4 per pound of $U_3O_8$ contained. Ore buyers in Colorado and Utah were offering 30 cents per pound for $V_2O_5$, and 55 cents per pound for $U_3O_8$ contents on the basis of about 4 per cent vanadium and 3 per cent $U_3O_8$. This amounted to only $57 a ton, and proved too low to encourage production. Inquiries for ore for export to Europe were made on the basis of 75 cents per pound of $U_3O_8$, f.o.b. New York. Since no payment was made for vanadium content this price was quivalent to only $45 for 3 per cent and $75 for 5 per cent ore. Evidently these prices were not enough to cover the cost of mining and transportation to New York and also provide a satisfactory profit, because little or no business seems to have been transacted.

Undoubtedly, a firm offer of any reasonably dependable tonnage of 5 per cent ore would develop a somewhat higher price. Several European firms are eager to get regular supplies, and probably would pay $1 or more a pound if they were assured of continued shipments.

The ore produced by the Bureau of Mines in 1917 cost $96 per ton, or $17 per milligram of radium recovered therefrom. The claims operated by the Bureau of Mines were probably at least as good as the average, and the costs of mining probably have not diminished since 1915. The general level of wages and commodity prices has increased roughly 50 per cent during the interval, and in mining small and erratic ore bodies there is little opportunity to offset wage increases by labor saving equipment and better mining technique. In 1918 carnotite containing a minimum of 2 per cent $U_3O_8$ was quoted at $2.75 to $3 a pound of uranium acid content; in July, 1922, while the American radium industry was still active, the quotation was $3.50. Several apparently well-informed engineers and prospectors are of the opinion that it would be necessary now to pay at least $2, and probably $3 or more per pound of $U_3O_8$ content in order to maintain a steady flow of carnotite ores from the Colorado-Utah fields. A price of $3 per pound of $U_3O_8$ is equivalent to approximately $23.50 per milligram of radium contained.

## Cost of Chemical Treatment

Exclusive of the cost of the ore, it cost the Bureau of Mines in 1915, $20.71 per milligram to extract radium. On the basis of present-day prices, this cost would doubtless be increased, and it should be remembered that the salaries paid the technical employees of the Bureau of Mines, in 1915 as now, were substantially less than would have been demanded of a commercial concern. In round figures, the cost per ton of ore treated was $120. Since it probably costs about as much to put a low-grade ore through the chemical plant as it does a high-grade ore, any savings in ore cost that would result from mining low-grade material may be largely offset by the increased cost of extracting the radium therefrom. Preliminary mechanical concentration, likewise, though it may raise the yield of ore and thus reduce costs at the mine, tends to produce a fine-grained or slime product that is more difficult and costly to treat.

## Summary of Cost Data

The National Radium Institute, under the technical management of the Bureau of Mines, produced over 8 grams of radium at a total cost (exclusive of credits for vanadium and uranium produced as by-products) of $37.60 per milligram. Doerner has correlated this and other available cost data in the form of a table:

Costs and yields of vanadium and radium from carnotite 1/

|  | (1) | (2) | (3) | (4) | (5)1/ | (6)1/ | (7)1/ |
|---|---|---|---|---|---|---|---|
| $U_3O_8$ in ore or concentrates, per cent | 2.66 | 3.00 | 2.00 | 1.00 | 2.50 | 3.00 | 2.75 |
| $V_2O_5$ in ore or concentrates, per cent | 4.30 | 2/5.00 | 4.00 | 4.00 | 5.00 | 5.50 | 5.00 |
|  |  | Cost per ton |  |  |  |  |  |
| Development | $5.00 | - | $45.00 | $5.00 | $15.00 | $20.00 | $15.00 |
| Mining | 36.00 | $7.15 | 50.00 | 25.00 | 75.00 | 100.00 | 100.00 |
| Transportation | 35.00 | 35.00 | 35.00 | 35.00 | 35.00 | 35.00 | 35.00 |
| Milling | - | 20.40 | - | - | 15.00 | 20.00 | 25.00 |
| Overhead, royalty, etc. | 20.00 | 21.75 | 20.00 | 5.00 | 5.00 | 5.00 | 5.00 |
| Total cost of ore or concentrates | 96.00 | 84.30 | 150.00 | 70.00 | 145.00 | 180.00 | 180.00 |
| Treatment | 118.00 | 2/125.00 | 125.00 | 125.00 | 150.00 | 150.00 | 150.00 |
| Total | 214.00 | 209.30 | 275.00 | 195.00 | 295.00 | 330.00 | 330.00 |
|  |  | Recovery |  |  |  |  |  |
| Radium, milligrams | 5.7 | 2/ 6.7 | 4.45 | 2.00 | 5.56 | 6.7 | 6.1 |
| $V_2O_5$, pounds | 25.0 | 2/ 85.0 | 68.0 | 68.0 | 85.00 | 93.00 | 89.00 |
| $U_3O_8$, pounds | 45.0 | 2/ 50.0 | 34.0 | 17.0 | 42.00 | 46.00 | 44.00 |
| Cost per milligram of radium | $37.60 | $31.30 | $61.80 | $97.50 | $53.00 | $49.25 | $54.20 |

1/ Doerner, H. A., Possibilities of Production of Radium and Vanadium from Carnotite. Ind. and Eng. Chem., vol. 22, No. 2, Feb., 1930, p. 185.

2/ Costs per ton of concentrates. 3/ Assumed.

Column 1 summarizes the Bureau of Mines' work in 1915; column 2 is based upon a modified procedure involving the milling of low-grade ore (0.85% $U_3O_8$ average) obtained as a by-product in mining the shipping ore; column 3 is Barker's [79] estimate of $150 per ton for 2 per cent ore, and $125 for treatment to recover radium only. Column 4, which is a similar estimate based upon 1 per cent ore, serves to demonstrate the point that increased treatment costs may more than offset savings in mining costs in the case of low-grade ore. Columns 5, 6 and 7 are estimates based upon some commercial tests made by Doerner on American ores in 1921.

It should be emphasized that the figures in the table, with the exception of columns 1 and 2, are not to be taken as representing actual costs but are given to illustrate the relations between grade of ore, cost of mining, and cost of radium with special reference to the possibilities of concentrating low-grade ores.

Barker's estimates (column 3), however, attempt to show the probable cost of production of radium from a typical American ore. Assuming that it would cost $62 per milligram to cover the bare costs of mining and processing, Doerner (reference cited) considers that after adding selling costs and a legitimate profit a selling price of $100 is not unreasonable. From the fact that domestic production of radium ceased when the Belgian product came on the market at $70, he adduces evidence that Barker's estimate comes close to actual costs under the practice then in vogue. Austrian costs, pre-war, were said to be around $80, and it is reported that the Czechoslovakian Government has lost money on its radium business in recent years. The costs of Radium Belge are not revealed, but there is reason to believe that they are probably under $20 per milligram.

## Recovery of Uranium and Vanadium

Uranium, as is well known, is a universal constituent of radium ores, and hence can always be produced as a by-product of the extraction of radium. Previous to the discovery of radium, ores were treated for their uranium contents, but in recent years the quantity of uranium available in residues from radium extraction has far exceeded the amount that can normally be absorbed. Efforts to utilize uranium as an alloying agent in steel have not met with much success. According to many authorities the addition of uranium is actually detrimental to steel, and the most favorable opinion appears to be that even if uranium does impart valuable properties, other elements considerably cheaper and more concenient to use are quite as effective. Aside from the small consumption of uranium in laboratory chemicals, virtually the only commercial use is as a coloring agent in glass and in ceramic glazes, notably for wall tile. Uranium oxide and salts for such purposes are sold at prices up to $3, and even $5, but probably not more than $2 per pound can be obtained from consumers who buy relatively large quantities under contract. The total volume of this business in the United States is not definitely known, but it probably ranges between 50,000 and 100,000 pounds (expressed as $U_3O_8$) the annual consumption being womewhat higher since 1928, due to a vogue (which may be temporary) for yellow and brown glass tableware. Whether a larger market would be opened up as a result of reduced prices is a question.

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[79] Barker, Howard H., Experiments on the Extraction and Recovery of Radium from Typical American Carnotite Ores. Univ. of Missouri Bull. vol. 24, No. 26, Sept., 1923.

Although American ores contain the valuable element vanadium, it is not present in ores produced in the Congo or other commercial localities. In the United States sandstones are found which contain vanadium minerals in various proportions, come carrying only roscoelite, the vanadium mica containing little or no uranium and others carrying carnotite high in uranium and low in vanadium. Although the ores have seldom been evaluated on the basis of both the radium and vanadium contents, the presence of vanadium in American ores constitutes a feature that should not be overlooked. When made into ferro-vanadium, for which there is an active demand, vanadium is worth $3.15 per pound, and a fair value for its oxide ($V_2O_5$) ranges from only 65 or 75 cents up to $2.50 per pound, depending upon purity and the extent of the market. Since the advent of Belgian radium on the market some tonnage of American carnotite has been treated in the United States for vanadium alone, the radium and uranium contents being discarded in the residues. Doerner's work, at the Reno station of the Bureau of Mines, however, definitely indicated that an ore containing as much as 1 per cent of $U_3O_8$ might be utilized as a commercial source of radium, provided all of the ore costs and part of the treatment costs are covered by the value of the vanadium. Doerner has summarized this situation as follows:

> "At the present time, however, with the decreasing amount of American vanadium available, our metallurgical industries are again turning toward the carnotite deposits. Vanadium is again the chief product sought and radium is not even recovered as a by-product on account of the Belgian competition. This situation is deplorable when the limited resources of this most valuable of all elements are considered.
>
> "The usual, and probably the best, method of recovering vanadium from the vanidiferous sandstones (roscoelite and carnotite) is to roast the ore with about 10 per cent of common salt and 2.5 per cent of soda ash. The calcine is first leached with water and then with dilute sulfuric acid and vanadic acid or ferrous vanadate in precipitated from the extracted solutions. Any radium present in the ore will be left as a fine precipitate in the tailings. If these tailings are discarded, the water used to flush them out and subsequent weathering will disperse the radium beyond all possibility of recovery. If, however, the tailings are deslimed, and the slimes are dewatered and stored, the radium will be saved in a concentrated form at a cost which is an insignificant fraction of its potential value, provided the original ore contains as much as 1 per cent $U_3O_8$.
>
> "If such low-grade ores can be produced in sufficient quantity to permit efficient countercurrent leaching operations on a large scale, it is probable that treatment costs could be considerably lowered. However, until it is certain that such operations would not break the present price of radium, the large investment required for such an enterprize would not be justified."

## The Need for Radium

Cancer takes a frightful toll of human life. It has been said that 10 per cent of all persons that survive the age of 40 die of cancer. Whatever the actual figures may be, anything that can be done to reduce the number that die annually of cancerous growths is distinctly in the interest of humanity. To the extent that radium saves lives and tends to prolong the useful span of human activity, it is a boon to mankind, and as such it has come within the sphere of governmental activities in many countries. In France the government has established 15 radium centers in various parts of the country, and allocated to them 31.5 grams of radium (in addition to about 20 grams believed to be privately owned.) In Sweden the use of radium is highly organized under Government auspices. A strong movement was started in 1929 to acquire a larger supply of radium in England, where Governmental committee recommended acquiring 24 grams in addition to about 25 grams then available in the British Isles. According to R. B. Moore 80/ the supply of radium in the United States is probably not more than that in France, say 50 grams, and it is doubtful if the world's supply of radium for medical purposes is more than 250 grams and probably less. Undoubtedly, more than one-half of the world's output has been dissipated through luminous paint, especially during the World War.

The best medical opinion seems to be largely in favor of using radium in large dosages. Under such circumstances small amounts in the hands of individual physicians are deemed of little real use, and considerations of public welfare demand that substantially greater amounts of radium be concentrated at convenient centers where a competent staff of specialists and the various accessories of a well equipped radiological laboratory can be maintained. The number of such well-equipped treatment centers is necessarily limited even in so large a country as the United States, and eventually a time may come when no more radium can be utilized effectively for therapeutic purposes. With cancer patients dying by the hundreds of thousands each year in the civilized countries of the world there is reason to believe that the saturation point has not been reached as yet, but the possibility of glutting the market is a major factor in the future of the radium industry. Competition from mesothorium has tended to reduce the consumption of radium in luminous paints, and improved technique employing deep-penetrating X-rays may displace radiotherapy in certain of its indicated applications, and thus further reduce the ultimate quantity of radium that can be sold.

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80/ Moore, R. B., The Present Radium Situation. Min. and Met., February, 1930, vol. 11, No. 278, PP. 91-92.

## APPENDIX I
## LIST OF RADIOACTIVE MINERALS [1]

| Name | Composition | Percentage | | Properties, etc. | Occurrences |
|---|---|---|---|---|---|
| | | $U_3O_8$ | $ThO_2$ | | |
| Ampangabeite | Niobate of uranium; rich in iron; contains titanium and tantalum | 14-20 | 0.5-3.0 | Orthorhombic; red-brown; sp. gr. about 3.4 | Madagascar (Ampangabe) |
| Autunite[1] | Phosphate of uranium and lime | 60 | 0 | Orthorhombic, tabular crystals, generally in veins in granite; greenish to sulphur yellow; sp. gr. 3.5-3.9 | Portugal<br>France (Autun)<br>Tonkin Madagascar Union of South Africa<br>Australiz,etc. |
| Betafite[1] | Niobo-titanate of uranium and lime | 26-30 | 0.5-1.7 | Octahedral crystals; darkbrown, turning yellow in alteration; sp.gr.4 | Madagascar (Betafo) |
| Blomstrandite | Tantalate, niobate, titanate of uranium and lime; contains rare earths | 20 | 0 | Amorphous; dark-brown; sp. gr. about 4.7 | Sweden<br>Madagascar |
| Broggerite (Variety of uraninite) | Oxide of uranium, lead, thorium, etc. | 77 | 7-8 | Octahedral; black; sp. gr. 9 | Norway |
| Carnotite[1] | Vanadate of uranium and potash | 61-67 | 0 | Yellow powder; orthorhombic crystals; mixture of quartz sand in canary-yellow sandstone; sp. gr. 4.1 | Colorado<br>Australia<br>Utah |

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1 - Data principally from M. Curie, Le Radium et les Radio-Elements. Paris, 1925, pp. 112-115, and from private notes of F. L. Hess.

List of radioactive minerals—Continued

| Name | Composition | Percentage | | Properties, etc. | Occurrences |
|---|---|---|---|---|---|
| | | $U_3O_8$ | $ThO_2$ | | |
| Chalcolite (Torbernite) | Phosphate of uranium and copper | 55-60 | 0 | Tetragonal, tabular crystals, often replacing crystals of autunite; green; sp. gr. 3.5 | Portugal<br>Spain, France<br>Bohemia<br>Saxony<br>Cornwall, etc |
| Cleveite | Oxide of uranium, thorium, and other rare earths; rich in helium gas | 57-72 | 4.1 | Cubical crystals; brownish-black; sp. gr. 7.5 | Norway<br>Texas<br>Carolinas<br>Saxony |
| Curite | Hydrous lead uranate | 73(max. | | | Belgian Congo |
| Davidite | Complex; contains vanadium, chromium, uranium, etc. | 4.6 | | Black mineral occurring in bright grains and cubelike crystals in pegmatite veins. | Olary,<br>S. Australia |
| Ellsworthite | Hydrated metatitanoniobate of uranium, calcium, and iron | | | Yellow or brown optically isotropic masses in pegmatites | Hybla,<br>Ontario,<br>Canada |
| Euxenite | Niobo-titanate of uranium, rare earths, and lime | 3-19 | 0.5-6.3 | Orthorhombic; generally in masses; brownish-black; sp. gr. 4.5 to 5 | Madagascar<br>Brazil<br>Sweden<br>Norway<br>Ireland<br>Carolinas |
| Fergusonite | Niobate of yttrium, rare earths, lime; low in uranium | 2-9 | 0.5-5.6 | Tetragonal; vitreous fracture; brownish-black; sp. gr. 5 to 5.8 | Norway<br>Sweden<br>Madagascar<br>Greenland<br>Russia<br>Carolinas |

List of radioactive minerals—Continued

| Name | Composition | Percentage | | Properties, etc. | Occurrences |
|---|---|---|---|---|---|
| | | $U_3O_8$ | $ThO_2$ | | |
| Gummite (Product of the decomposition of uraninite) | Silicate of uranium, lead, lime, barium; | 60-70 | 0-0.3 | Amorphous; reddish yellow; sp. gr. 4 | Belgian Congo<br>Saxony<br>Bohemia<br>North Carolina<br>Tunis |
| Johannite | Sulphate of uranium and copper | 66 | 0 | Monoclinic; green | Bohemia<br>Saxony |
| Kasolite | Hydrous silicauranate of lead | | | | Belgian Congo |
| Liebigite | Uranium carbonate and calcium hydrate | 37-38 | 0 | Transparent | Austria<br>Saxony<br>Russia<br>Adrianople (Turkey) |
| Medjidite | Sulphate of uranium and lime | | | Transparent; dark yellow | Adrianople (Turkey) |
| Monazite[2] | Anhydrous phosphate of cerium earths; contains thorium | | 0.1-13.5 | Monoclinic, usually in alluvial deposits; sp. gr. 4.9 to 5.3 | Brazil<br>Carolinas<br>Northern Africa<br>India<br>Australiz<br>Russia<br>Norway<br>Madagascar,etc |
| Pitchblende[1] (Amorphous uraninite) | Oxide of uranium, lead; contains rare earths | 75-95 | 0<br>traces | Black; resinous luster; sp. gr. about 6.5 | Bohemia<br>St. Joachimsthal<br>Belgian Congo<br>Cornwall<br>Colorado<br>Saxony<br>Canada, etc. |

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[1] - Ore of radium and uranium.

[2] - Thorium ore.

List of radioactive minerals—Continued

| Name | Composition | Percentage | | Properties, etc. | Occurrences |
|---|---|---|---|---|---|
| | | $U_3O_8$ | $ThO_2$ | | |
| Polycrase | Columbo-titanate of rare earths (Y, Er, Ce) and Uranium | 6-19 | | Orthorhombic; crystals thin, prismatic, tabular; cleavage none; fracture conchoidal | Hitterö, Norway; Slaettakra, Alsheda, Smaland, Sweden; near Dresden; Henderson County, N.C.; Greenville County, S.C.; Twp. of Calvin, Nipissing, Ontario, Canada |
| Samarskite | Niobo-tantalate of uranium, rare earths, and lime | 3-16 | 0.5-4.5 | Orthorhombic; vitreous; brownish-black; sp. gr. 4.4 to 5.7 | Sweden<br>Norway<br>Carolinas<br>Colorado<br>Canada<br>Madagascar |
| Samiresite | Variety of betafite, containing considerable lead | | | | Madagascar (Samireay) |
| Thorianite | Oxide of thorium, uranium, and rare earths | 5-12 | 68-73 | Cubiczl crystals; black; sp. gr. 9.3 | Ceylon |
| Thorite | Silicate of thorium and uranium | 1-22 | 45-57 | Tetragonal; in brownish masses; sp. gr. 4.8 to 5.4 | Norway<br>New York |
| Torbernite (See Chalcolite) | | | | | |

List of radioactive minerals—Continued

| Name | Composition | Percentage | | Properties, etc. | Occurrences |
|---|---|---|---|---|---|
| | | $U_3O_8$ | $ThO_2$ | | |
| Torbernite (See Chalcolite) | | | | | |
| Troegerite | Uranium arsenate, hydrated | 66.3 | 0 | Monoclinic tabular crystals; lemon-yellow | Saxony |
| Tyuyamunite | Hydrous uranovanadate of Ca | 59 | | | Tyuya-Muyun, Fergana, Russian Central Asia |
| Uraninite | Uranium oxide, lead, rare earths; contains thorium | 75–95 | 0.5–11 | Octahedral; conchoidal fracture; brownish black; sp. gr. 9 to 9.7 | Norway<br>Carolinas<br>Texas<br>Connecticut<br>Eastern Africa<br>Saxony<br>Spain, etc. |
| Uranophane | Hydrous silicate of uranium and calcium | 52–65 | | Orthorhombic; minute acicular prisms in radiated or stellate aggregations; also massive with fibrous structure | Kupferberg, Silesia; Wölsendorf, Bavaria; Neustaedtel, Saxony; Mitchell Co., N. C.; Ottawa Co., Quebec; Stone Mountain, Ga. |

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1 - Ore of radium and uranium.

2 - Thorium ore.

## APPENDIX II

## RADIOACTIVE CONSTANTS

### Atomic and Molecular Weights

Radium (Ra) 225.95; Uranium (U) 238.17; Vanadium (V) 50.96; Oxygen (O) 16; Chlorine (Cl) 35.46; Bromine (Br) 79.92; $U_3O_8$ 842; $V_2O_5$ 182; $RaBr_2.2H_2O$ 422.

### Carnotite Yield Ratios

Radium-uranium ratio = 3.40 x $10^{-7}$

1 short ton $U_3O_8$ contains 1,697 pounds or 769.75 kg. V.

1 kg. (2.2046 lbs.) U contains about 1/3 mg. Ra.

1 short ton U contains 302.4 mg. Ra.

1 short ton $U_3O_8$ contains 256.58 mg. Ra.

1 short ton 1 per cent $U_3O_8$ contains 2.5658 mg. Ra. of which say 90 per cent (2.309 mg.) may be recoverable, hence about 215 tons of 2 per cent $U_3O_8$ or 145 tons of 3 per cent $U_3O_8$ ore are required to furnish 1 gram of Ra.

Uranium-vanadium ratio in carnotite or tyuyamunite (theoretical) = 3.09

1 kg. $V_2O_5$ contains 0.5602 kg. V.

1 short ton 1 per cent $V_2O_5$ contains 11.20 lbs. V.

### Radium Constants[1]

| | |
|---|---|
| Half-decay period | 1690 years[2] |
| Transformation constant | 4.38 x $10^{-4}$ per year |
| Type of radiation emitted | Alpha |
| Range of rays in air S. T. P. | 3.21 cm. |
| Current produced by alpha particles from 1 g. radium (without equilibrium products) | 2.41 x $10^6$ e. s. units |

### Radium Emanation Constants[1]

| | |
|---|---|
| Atomic weight | 222 |
| Half-decay period | 3.81 d. |
| Transformation constant, | $0.18\ d^{-1}$ |
| or | $0075\ hr.^{-1}$ |
| Type of radiation emitted | Alpha |
| Range of rays in air | 3.91 cm. |
| Current produced by alpha particles from per Curie emanation alone | 2.75 x $10^6$ e. s. units |

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[1] - Taken from Barker, Howard H., and Schlundt, H., Experiments on the Extraction and Recovery of Radium from Typical American Carnotite Ores Univ. Missouri Bull., vol. 24 No. 26, Sept., 1923, pp. 80-4. Substantially different figures for the rate of transformation are given by later writers (e. g. Robertson and Miller, quoted on p. 14 of this paper.

[2] - Barker and Schlundt give 1580.

## Absorption of Various Types of Rays[1]

| | |
|---|---|
| Alpha rays: | |
| Completely absorbed by | .006 cm. of Al foil |
| Hard beta rays: | |
| Completely absorbed by | 2 mm. of lead |
| Half absorbed by | .1 mm. of lead |
| Hard gamma rays: | |
| Half absorbed by | 14 mm. lead |

## Miscellaneous

| | |
|---|---|
| Charge carried by alpha particle | $9.54 \times 10^{-10}$ e. s. units |
| Number of alpha particles expelled per second per gram of radium itself | $3.72 \times 10^{10}$ |
| Number of alpha particles expelled per second per gram of radium in equilibrium with its products, inclusive of RaC | $14.88 \times 10^{10}$ |
| Total number of pairs of ions produced by one alpha particle from RaC in air by complete absorption | $2.20 \times 10^5$ |
| Velocity of alpha particle from Ra C' | $1.922 \times 10^9$ cm. per sec. |
| Velocity of alpha particles in miles per second | 9,000 to 12,000 |
| Velocity of hard beta rays, about | $2.9 \times 10^{10}$ cm. per sec. |
| or | 170,000 miles per sec. |
| Velocity of gamma rays | 180,000 miles per sec. |
| Helium generated per year per gram of radium | 174 $mm.^3$ |
| Quantity of heat liberated by 1 gram of radium and its equilibrium products per hour | 136 cal. |
| Distributed as follows: | |
| Alpha rays | 125 cal. |
| Beta rays | 4.5 cal. |
| Gamma rays | 6.5 cal. |
| Volume of 1 curie of emanation | 0.663 $mm.^3$ |
| Weight of 1 curie of emanation = $6.56 \times 10^{-6}$ g. = | 6.56 micrograms |
| Ionization due to alpha, beta, and gamma rays is of the relative order of | 10,000, 100 and 1 |
| Velocity of alpha particle | $1.0246 \times 10^9 R_0^{1/3}$ |
| Number of pairs of ions produced in air by any alpha particle | $6.25 \times 10^4 R_0^{2/3}$ |

$R_{15^\circ} = 1.055 R_0$; $R_{20^\circ} = 1.073 R_0$

1 Mache unit represents $3.64 \times 10^{-10}$ curie per liter

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1 - Taken from Barker, Howard H., and Schlundt, H., Experiments on the Extraction and Recovery of Radium from Typical American Carnotite Ores. Univ. Missouri Bull., vol. 24, No. 26, Sept., 1923 pp. 80-4. Substantially different figures for the rate of transformation are given by later writers (e. g. Robertson and Miller, quoted on p. 14 of this paper).